An Overview of Sediment-Associated Contaminants and their Bioassessment

M. Munawar[*], R.L. Thomas[*], H. Shear,
P. McKee and A. Mudroch.

[*] Great Lakes Fisheries Research Branch
Department of Fisheries and Oceans
Canada Centre for Inland Waters
P.O. Box 5050, Burlington, Ontario L7R 4A6

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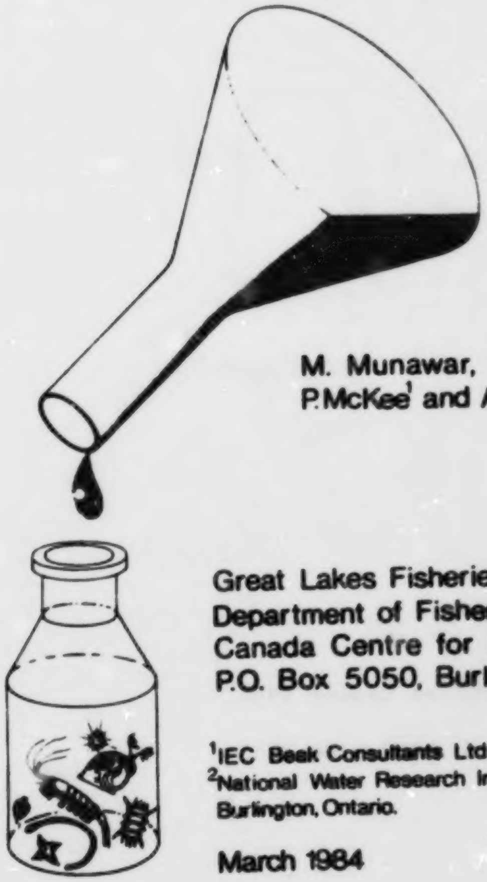

M. Munawar, R. L. Thomas, H. Shear,
P. McKee[1] and A. Mudroch[2]

Great Lakes Fisheries Research Branch
Department of Fisheries and Oceans
Canada Centre for Inland Waters
P.O. Box 5050, Burlington, Ontario L7R 4A6

[1] IEC Beak Consultants Ltd., Mississauga, Ontario
[2] National Water Research Institute,
Burlington, Ontario.

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ABSTRACT

Munawar, M., R.L. Thomas, H. Shear, P. McKee and A. Mudroch. 1984. An overview of sediment-associated contaminants and their bioassessment. Can. Tech. Rep. Fish. Aquat. Sci., 1253 pages i - vi, 1 - 136.

An overview of the published material concerning sediment contamination by trace metals and persistent organics is presented. Information from the Great Lakes is highlighted. Physico-chemical processes governing the behaviour of contaminants in sediments are discussed. The impact of sediment contamination on aquatic biota as well as effect of biota on contaminants are considered. Approaches and procedures used to determine the bioavailability and bioassessment of sediment-associated contaminants are outlined.

RESUME

Munawar, M., R.L. Thomas, H. Shear, P. McKee and A. Mudroch. 1984. An overview of sediment-associated contaminants and their bioassessment. Can. Tech. Rep. Fish. Aquat. Sci., 1253 pages i - vi, 1 - 136.

Dans cet article, on présente une vue d'ensemble des travaux publiés sur la contamination des sédiments par des métaux en traces et des produits organiques persistants. On met en relief l'information relative aux Grands Lacs. On examiné les processus physico-chimiques qui régissent le comportement des contaminants dans les sédiments. On considère l'incidence de la contamination des sédiments sur les biotes aquatiques ainsi que l'effet des biotes sur les contaminants. On présente les méthodes et procédés employés pour déterminer la biodisponibilité et la bioévaluation des contaminants associés aux sédiments.

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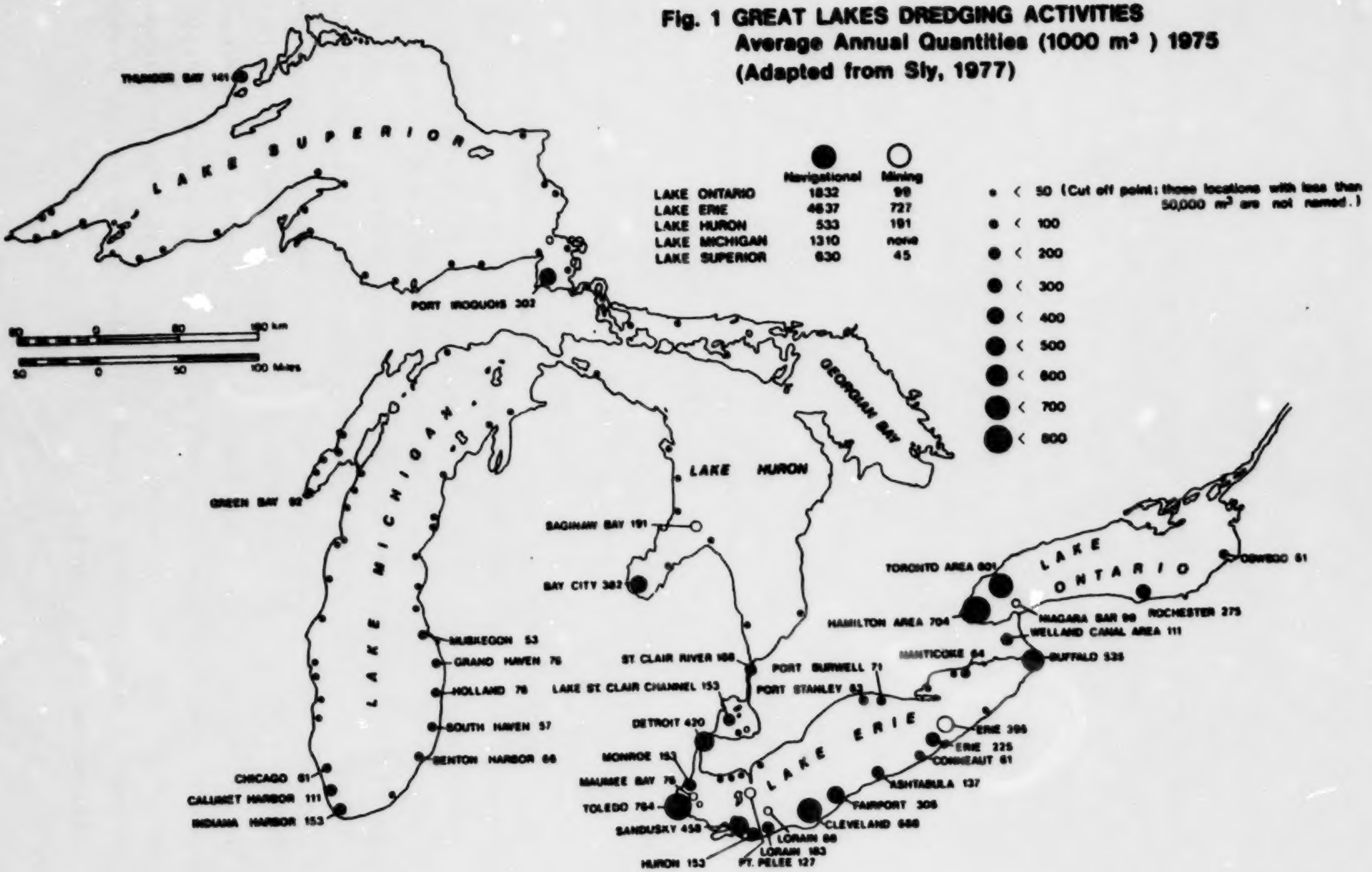
INTRODUCTION

Recent changes in the environment and in the biological communities of aquatic ecosystems are directly or indirectly attributable to anthropogenic loading of contaminants. Many contaminants, including trace contaminants and persistent organic compounds, show acute and chronic toxicities to aquatic organisms and may pose serious health hazards to humans. The toxicity to an ecosystem will depend on the availability of trace metals for metabolic processes which, in turn, are closely related to their chemical species both in solution and in particulate matter. For this reason a number of investigations on the behavior of trace metals in the dissolved state were undertaken (Fitzwater et al., 1982; Damielsson, 1982). In comparison, very little is known about the chemical association of metals in suspended materials and sediments, and their availability for metabolic processes. Generally, even less is known about the behaviour and associations of organic contaminants in aquatic ecosystems.

Recently, dredging operations have become a major concern in the Great Lakes nearshore zone. Dredging operations and dredged material disposal may effect water and sediment quality. This re-distribution and transportation of contaminated sediment from areas such as harbours and shipping channels to less contaminated disposal sites re-mobilizes sediment-bound contaminants. The significance of the environmental impacts of dredging and disposal operations was recognized in the 1972 Great Lakes Water Quality Agreement. Later, during 1972, an International Working Group on the abatement and control of pollution from Dredging Activities was established in compliance with Article V - Section (f) of the Agreement. It was determined that over ten million cubic meters of sediment are dredged annually in the Great Lakes and approximately one half of the dredging is occurring in Lake Erie. It has been estimated that more than 90 percent of dredging is associated with the maintenance of harbours and waterways for navigational purposes (Report to the Great Lakes Water Quality Board by the Dredging Subcommittee - 1982, see Fig. 1).

Fig. 1 GREAT LAKES DREDGING ACTIVITIES
Average Annual Quantities (1000 m³) 1975
(Adapted from Sly, 1977)



This overview is intended to provide the present state-of-the-art concerning the sediment-associated contaminants in relation to the bioassessment procedures. Furthermore, the overview will hopefully assist in the development of guidelines for the disposal of dredged material in the Great Lakes and facilitate the modification of the register for the evaluation of Great Lakes dredging activities by the Dredging Subcommittee, Water Quality Board, 1982.

SEDIMENT CONTAMINATION BY TRACE ELEMENTS

Research on trace metal pollution of aquatic environments has been extensive, particularly since the Minamata (Japan) tragedy in 1953 in which 41 people died from consuming mercury-contaminated seafood (Kurtland et al. 1960). Aquatic ecosystems consist of three reservoirs for trace metals: water, sediment, and biota. Metal concentrations in these reservoirs are determined by a complex equilibrium of various physical, chemical and biological factors.

Walters et al. (1974) listed four stages in the geochemical cycle of trace metal pollutants in aquatic environments: 1) introduction into the aquatic environment, 2) physical transport and dispersal within the basin, 3) transfer out of the water mass, mainly through sedimentation and burial, but also to some degree by vaporization losses to the atmosphere, and 4) post-depositional transfer of buried pollutants back across the sediment-water interface into the overlying hydrosphere. It is the purpose of this section to summarize the literature concerning the biogeochemical processes governing the behaviour of trace metals in the aquatic environment.

Sediment-Water Interchange

A large amount of recent research on sediment-water interchange of trace metal contaminants has evolved from the establishment of legislation designed to minimize environmental impact from contaminants released during dredging operations in the United States. The National Environmental Policy Act of 1969 requires detailed environmental impact assessments of proposed new navigational dredging projects and projects requiring maintenance dredging. The River and Harbor Act of 1970 authorizes a "comprehensive program of research, study, and experimentation relating to dredged spoil".

Trace metals occur in many different forms in the aquatic environment. Metals may be present in solution from the dissolution of minerals during natural erosion and from the discharge of mining and industrial wastes. Metals may then be adsorbed on, co-precipitated with, or occluded within various inorganic and organic matrices. The settling of this solid suspended matter transports metals from the aqueous phase to the sediments.

Sediments generally act as a reservoir for trace metals in aquatic environments. For instance, most Hg (up to 97 percent) found in the Ottawa River is associated with bed sediments (Kudo et al. 1977a). Data of DeGroot and Allersma (1975) and DeGroot (1976) on European rivers indicated that most heavy metals transported in river water are associated with suspended sediments. In Indian Lake, Massachusetts, 98 percent of the Cu retained by the system following a copper sulphate treatment was present in the sediment and only about one percent of the total sediment Cu load could be removed by washing with deionized water (Symmes 1975). Studies described by Hetherington and Harvey (1978) demonstrated that 95 percent of radioactive Pu discharged into seawater is lost to the sediment phase almost "immediately" and that other nuclides, particularly dissolved lanthanides and actinides, behave similarly.

Sediments in the Great Lakes basin also are important reservoirs for trace metals. Fitchko and Hutchinson (1975) examined the sediments deposited at the mouths of 116 tributaries flowing into the Great Lakes and found elevated concentrations of Pb, Ag, Cd, Co, Cu, Cr, Ni, Zn, Mn and Hg in sediments of streams draining industrialized areas. Exceptions occurred where swamps and small glacial lakes act as sedimentary sinks between contaminant sources and stream mouths. High Ni and Cu concentrations were found in the sediments of a Lake Huron tributary draining a portion of the metal-contaminated Sudbury smelting area (Hutchinson et al. 1976). Enrichments of several heavy metals due to anthropogenic loadings have been found in the sediments of the St. Clair River and the Detroit River (Environmental Control Technology Corporation 1974). The International Joint Commission (1978) showed that Pb and Hg levels in the bottom sediments of the Great Lakes are closely correlated with river inputs of these metals and that sediment metal concentrations and distribution patterns reflect river and lake transport mechanisms. Elevated concentrations of trace metals in surface sediment relative to subsurface sediment have been found in Lakes Superior (Nussman 1965; Helmke et al. 1977; Kemp et al. 1978), Michigan (Shimp et al. 1970; Edgington and Robbins 1976), Huron (Thomas 1973; Kemp and Thomas 1976a; Kemp et al. 1978), Erie (Walters et al. 1974; Kemp et al. 1976) and Ontario (Thomas 1973; Kemp and Thomas 1976) due to anthropogenic metal inputs.

Metal-Sediment Associations

Förstner (1977) described a variety of geochemical metal associations in sediment:

1) Heavy metals are transported and deposited as constituents in the natural inorganic detritus, mostly in inert positions

2) Slightly alkaline waters promote the formation and precipitation of hydroxides, carbonates, sulphides and phosphates of heavy metals

3) Sorption and cation exchange occurs on fine-grained particles with large surface areas. The general sequence of the sorptive capacity of solids is MnO_2 >humic acids >hydrous Fe oxides >clay. The sorptive capacities of different clays also vary (Buorg and Filby 1974; Farrah and Pickering 1977)

4) Dissolved organic compounds complex with dissolved metals. Less condensed humic acids (fulvic acids) are particularly important in complexing with heavy metals due to a large number of functional groups on organic molecules. While contamination by heavy metals may be correlated with high sediment organic levels, this does not always indicate direct bonding of metals to organics. In highly polluted areas, these correlations may be largely coincidental due to urban, industrial or agricultural waste loadings of both metals and organic contaminants

5) Heavy metals may co-precipitate with hydroxides and carbonates. Heavy metal carbonates having low solubilities will be completely

precipitated from solution as a result of co-precipitation with calcium carbonate. Iron hydroxide concentrations can have metal enrichment factors of 30 times for Cd, 17 for Zn and 1.4 for Cu in comparison with clays in the surrounding sediments. Similarly, calcium carbonate coatings in river sediment can have enrichment factors of 10 times for Cd, 6 for Cu, 4.5 for Zn and 4 for Co when compared to geochemical background concentrations in carbonate rocks.

Sorption-Desorption

Processes controlling sorption and desorption determine the sediment-solute distribution of trace metals. A literature review on release and adsorption of metals by freshwater sediments was provided by Cavagnaro (1980). Two of the most important sediment characteristics governing sorption and desorption of metals are particle size distribution and organic matter content. Ramamoorthy and Rust (1976) and Townsend et al. (1974) found that grain size and organic content determined the quantity of Hg sorbed by Ottawa River sediments. Sediment concentrations of Cu, Fe and Zn correlated well with percent clay and organic content of Lac St. Jean sediments (Ouellet 1979). DeGroot and Allersma (1975) demonstrated that sediment levels of several heavy metals increased with increasing percentage of <16 μm particles (medium silt) in the Ems River, Germany. Förstner et al. (1979) and Grieve and Fletcher (1976) documented distinct reductions in metal content with increasing grain size in Rhine River (Germany) and Fraser River (British Columbia) sediments, respectively. Rickert et al. (1976) distinguished between background concentrations and polluted conditions for Cr, Cu, Pb, Hg, and Zn in sediments of the Willamette River, Oregon, by analyzing conditions in the <20 μm fraction. Jackson (1977) found strong correlations between organic carbon and levels of Hg, Cu, Zn, Cd and Fe in Clay Lake and Ball Lake, Ontario. Lindberg and Harriss (1974) demonstrated the association of Hg with organic matter in the Florida Everglades and in the Mobile Bay estuary. Sediment Hg was shown to correlate with percent clay content, but not with organic carbon in an Italian Lake (Damiani and Thomas 1974).

Similarly, relationships between metal levels and the parameters of grain size and organic content in the Great Lakes basin have also been studied. A number of significant correlations of heavy metals with clay and organic matter content were documented for sediments from the Great Lakes and from the mouths of tributary streams (Hutchinson and Fitchko 1974; Fitchko and Hutchinson 1975). Negative correlations were found between Cu, Zn, Cd, Cr, Ni and Hg levels and sand content and positive correlations between the same metals and silt content in sediments at the mouth of the Fox River (Wisconsin) into Lake Michigan (Pezzetta and Iskandar 1975). In an acid-peroxide extract of Lake Michigan sediments, concentrations of Cu, Fe and Zn correlated well with grain size (Baker-Blocker et al. 1975). Owen (1979) examined relationships between levels of Fe, Cr and Cu and grain size in sediments of Little Traverse Bay, Lake Michigan and found that concentrations in the fine-grained fraction were not proportional to the increased surface area of the grains when compared with larger grain size fractions. Levels of Hg in Lake Erie sediment samples were found to correlate with organic carbon content but not with percent clay (Thomas and Jaquet 1976). The relationship between Hg content and organic matter was demonstrated by Kennedy et al. (1971) and Cline et al. (1973) for Lake Michigan and St. Clair River sediments, respectively. Metal levels were related to percent organic matter in suspended sediment in Wilton Creek, a Lake Ontario tributary (Ongley 1978). Activity and isotopic ratios for Pu-238, Pu 239, 240 and Cs-137 were determined for Lake Michigan sediments by Albert and Muller (1979) who found that the distribution of isotopes was unrelated to particle size.

Grain size and organic content may be considered physical properties of sediment. Chemical properties governing the sorption and desorption of trace metals include salinity, pH, Eh, and the presence of complexing agents.

Increased salinity results in competition between dissolved cations and adsorbed metals, resulting in partial displacement of the latter (Förstner 1977). This process may be complicated by a decrease in sediment-bound heavy metals due to mixing of polluted river deposits with relatively uncontaminated marine sediment. The displacement of sediment-bound heavy metals by an elevation in salinity is likely of minor importance in the geochemistry of Great Lakes sediments.

The literature dealing with the role of pH in trace metal chemistry was reviewed by Khalid et al. (1977). A decrease in pH leads to the dissolution of metal carbonates and hydroxides and to increased desorption from electrostatic associations with sediment particles. At low pH's protons compete for adsorption sites on sediment particles. Payne and Pickering (1975) found that kaolinite removes all Cu (II) from solution at pH>6 while at pH<3.5, no Cu uptake by clay occurred due to competition with H^+. Similar observations were made by Farrah and Pickering (1977) and Frost and Griffin (1977) who studied the removal of various heavy metals from solution by clay suspensions. Ramamoorthy and Rust (1976) have shown that the rapid transfer of Hg to Ottawa River sediments is little affected by pH. Khalid et al. (1979) demonstrated that Cd in estuarine sediment is transformed into more mobile, soluble forms as pH declines. Huang et al. (1974) studied the effects of pH on sediments from the Missouri River watershed and found that a pH increase promotes the dissolution of organic matter and associated contaminants, including metals, from the solid phase. Allin (1978) raised concern that the increasing acidity of precipitation may lead to the release of sediment-bound toxic metals. Schindler et al. (1980) observed the behaviours of metal radioisotopes during acidification within enclosures in a Shield lake. A decrease in pH from 6.7 - 6.8 to about pH 5 increased the solubilities of Fe, Co, Mn and Zn but decreased the solubilities of V and Hg. Acidification also slowed the rate of loss of Mn and Zn but increased the loss of Ba, Se, Cs and V to the sediment. Furthermore, acidification induced the release of Al, Mn, Zn and Fe from the lake sediment. It is evident that pH plays an important role in sediment metal chemistry, although the effects of pH on sorption and desorption vary with different metal and sediment properties.

Khalid et al. (1977) also reviewed the literature concerning metal transformations as affected by oxidation-reduction. Changes in redox conditions are frequently the result of changes in dissolved oxygen concentrations owing to organic decomposition. Oxygen deficiency leads first to the dissolution of hydrated manganese oxide and then to the dissolution of hydrated iron oxide. Tessenow and Baynes (1975) and Michard (1975) concluded that remobilization of Fe and Mn in anoxic sediments followed by upward diffusion may account for accumulations of Fe and Mn oxides at the oxidizing-reducing interface. Since Fe and Mn are soluble in their divalent states, other metals co-precipitated with Fe and Mn oxides tend to be remobilized under anoxic conditions. Reinhard and Förstner (1975) demonstrated that Cu, Zn and Cd may be released by anoxic sediments into over-lying water, although it has been suggested that heavy metal release under anoxic conditions may be attributable more to bacterial activity rather than to ion migration (Cline and Upchurch 1975).

Opposing the tendency of heavy metals to be released by oxides of Mn and Fe under reducing conditions is the process of metal reaction with sulphides to form stable compounds in sediments. Many heavy metals such as Hg, Zn and Cu are firmly bonded and precipitated as sulphides and may, therefore, accumulate in sulphide-rich sediments where organic decomposition has led to reducing conditions (Allin 1978). Subsequent oxygenation of the sediments or the addition of natural oxidants such as ferric iron or exotic oxidants such as chlorine may cause metal release into the aqueous phase (Bolshakov 1969). Jackson (1978) proposed that sewage be introduced with metal-contaminated effluents in tailings ponds at Flin Flon, Manitoba, since the subsequent production of sulphide would lead to entrapment of metals in the sediments.

Complexing agents such as humic acids, amino acids, ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA) and sodium tripolyphosphate (TPP) compete with sediments for dissolved metals. The presence of complexing agents can mobilize major cations and some heavy metals from bottom materials into solution (Chau and Shiomi 1972; Barica et al. 1973; Lahar and Hochberg 1975; Inoue and Suemitsu 1979). The growing use of complexing agents (such as NTA) in detergents may increase the dissolution of heavy metals from aquatic sediments (Chau and Shiomi 1972).

Conversely, complexing agents may mediate the sorption of heavy metals by sediments under some conditions. The existence of ternary compounds of surface-metal-ligand and surface-ligand-metal has been demonstrated in several systems (Bourg 1979). Depending on the number and type of ligands involved, complexing agents may bond both with metals and solid surfaces (Elliott and Huang 1980).

The nature of metal-organic interactions is complex. A major problem in the study of these interactions is the fact that of the organic species involved, humic acids, fulvic acids and humins are the most important and form part of an extremely heterogeneous polymer system (Jonasson 1977). Differences in elemental composition, acidity, degree of polymerization and molecular weight all affect the nature of organic-metal associations. Jonasson (1977) reviewed information on organic-metal interactions in detail.

The relative affinities of metals for different binding agents vary systemmatically with metal properties (Jackson 1979). Walters et al. (1974) related enrichment factors of metals in surface sediment relative to subsurface sediment in Lake Erie to ionization potential, with metals having higher ionization potentials being selectively incorporated into sediments due to their bonding properties.

Jackson et al. (1980) related metal-sediment associations in a Shield lake to metal properties. Metals of high stabilization energy, high electronegativity, or small ionic radius were most readily adsorbed by suspended particles and colloids and were preferentially adsorbed by sediment organic and inorganic binding agents. The authors showed that factors such as pH and the type of binding agent governed which metal properties exerted the dominant effect on metal behaviour. For instance, Hg was adsorbed quickly at pH 6.7 to 6.8 due to its high electronegativity, but was removed more slowly from solution than other metals at pH 5.1 owing to its large ionic radius. Metals also displayed increasing affinities for high molecular weight humic materials with increasing electronegativity so that Se and Hg were more strongly bonded to humic substances than were other metals.

A review of 227 papers on interactions between metals, sediments and water was presented by Jackson et al. (1978). Topics covered included metal and organic speciation studies; sorption-desorption interactions with organic matter, clay minerals and humic acids; chemical reactions between humic acids, heavy metals and silicate minerals; oxidation-reduction reactions between metal ions and humic acids; and effects of sulphides on some sediment-metal interactions.

A few workers have developed models of heavy metal exchange between sediment and water. Baccini et al. (1977) used a thermodynamic approach by considering the formation of certain stable metal species in a set of possible inorganic and organic ligands. The proposed model treated plankton, allochthonous particles and unidentified dissolved substances as uniform ligands under a given set of conditions. Lu and Chen (1977) set up a model for metal solid-solute equilibria controlled by sulphides and found good agreement between model calculations and experimental results using three types of sediments. Based on published information and models dealing with the distribution of heavy metals in the aquatic environment, Jorgensen (1979) developed differential equations to model heavy metal exchange between sediment and water.

Elemental Interrelationships

The literature on interrelationships of elements in sediments was reviewed by Leland et al. (1977). Cronan and Thomas (1972) studied the geochemistry of ferromanganese oxide-coated sands and manganese nodules in northern Lake Ontario. Levels of Mn, Ni, Co and Zn showed negative correlations with Fe content. Black, Mn-rich coatings on rocks in northern western United States streams were enriched with Zn, Ni and Co, occassionally with Cu, but never with Pb relative to 80 µm sediments (Carpenter et al. 1975). In the Thames River estuary (England) total Fe in sediment was correlated positively with Ce and Co but negatively with Zn (Bevan et al. 1975). Loring (1976) noted an association of acid-soluble Cu and Zn with sedimentary Fe while total Pb was related to Fe, Mn and carbonate in sediments of the Saguenay fjord. In lakes near Sudbury, Ontario, correlations of C, Fe and Mn with sediment chemical properties were overshadowed by rates of Cu, Ni, Pb and Zn deposition.

Rust and Waslenchuk (1974) found that Hg, Mn and Fe concentrations in surface coatings on sediment grains in the Ottawa River were interrelated with a Hg:Mn:Fe ratio of about $1:10^3:10^4$. Although inter-element correlations are common in sediments of mixed grain size, some correlations may disappear when comparisons are made within sediment size fractions. For instance McDuffie et al. (1976) found no correlation of Ag, Cd, Cu, Ni or Zn with either C or Mn within grain size fractions in the Susquehanna River, although Zn and Ni were correlated with Fe in the clay fraction.

Relationships among elements have been used to develop indicators of depositional factors. Baker-Blocker et al. (1979) correlated levels of Cu, Fe and Zn in surface sediments of Lake Michigan with grain size. A high metal content relative to grain size in sediments of a tributary demonstrated that the river was a source of metal contamination in the lake. Plato and Jacobson (1976) found Cs-137 level to be a good indicator of Pu-238, 239, Zn, Cu, Cr and Pb but a poor indicator of Sr-90, Ni and pesticides in Lake Michigan sediments. The ratios of Zn:Mn and Ni:Mn were used to separate areas of Zn and Ni mineralization (high ratio) from areas of co-precipitation (low ratio) in a Canadian Shield lake (Coker and Nichol 1975). Copper enrichments in Newfoundland inlets were recognized by deviations from the normal high correlation between Cu level and percent clay (Slatt and Sassville 1976). The ratio of Co-60:Cs-137 was used as an indicator of Co-60 deposition from a Japanese nuclear power plant (Nagaya and Nakamura 1975). Thompson and Paton (1975) used Cu:Zn ratios as an indicator of Cu deposition into sediment from mine tailings. Leland et al. (1977) pointed out that both depositional and physicochemical causes of elemental interrelationships must be given consideration in the interpretation of such data.

SEDIMENT CONTAMINATION BY PERSISTENT ORGANICS

Research on persistent, toxic organic compounds in the aquatic environment is relatively recent. It was not until the late 1960's and early 1970's that the combination of increased environmental awareness and laboratory analytical capability sparked research on the occurrence and effects of pesticides and related substances in the aquatic environment.

The Research Advisory Board of the International Joint Commission (1978) compiled a list of approximately 2,800 synthetic chemicals produced or used within the Great Lakes basin. Many of these compounds pose environmental and human health threats and some persist in the environment. Organochlorines are generally more persistent and have attracted more attention than any other group of chemicals.

Recently, the shift in pesticide use in the Great Lakes basin has been from persistent pesticides of relatively low toxicity to those of much less persistence but often of greater toxicity (Pionke 1977). Three pesticidal properties are of primary significance: persistence, toxicity, and the pesticide distribution between phases of the aquatic environment (adsorptivity). Pionke (1977) made the following observations on these three basic properties. These observations on pesticides also pertain to related compounds not used for pest control, particularly polychlorinated biphenyls (PCB's).

The persistence times (half-lives) can be divided into three groups: $\leq$ 6 months, 6 to 18 months and $\geq$ 18 months. However, the International Joint Commission (1975) defined a persistent compound as one which either a) by itself or as its transformation product has a half-life for degradation under natural environmental conditions of more than eight weeks, or b) by itself or as its transformation products may bioconcentrate in the biota of the receiving system. Short-lived compounds usually exert more localized environmental impacts. The most persistent compounds exert their influence over considerable distances and tend to accumulate in the aquatic environment. Compounds of intermediate persistence vary in their environmental impact, depending upon the formation of toxic daughter compounds and the possibility of an increase in persistence under certain environmental conditions.

The second property following persistence is toxicity. While acute toxicities of most pesticides are known, little knowledge exists on chronic toxicity and on synergistic effects of pesticide combinations. Pionke (1977) summarized information on persistence and toxicity of pesticides currently in common use in the Great Lakes basin.

The third important pesticidal property is adsorptivity. Adsorption of organic compounds onto particulate matter tends to increase persistence and to decrease bioactivity. Pesticide and sediment properties influencing the adsorption process are described in a subsequent section (Impact of Metal-Contaminated Sediment on Biota).

Persistent organic compounds are represented by pesticides (primarily herbicides, insecticides and fungicides) and related compounds such as polychlorinated biphenyls (PCB's). Due to the molecular complexity of most of these organic contaminants, chemical nomenclature is also generally complex. Throughout the following discussion, compounds will usually be referred to by their commonly used, abbreviated names or trade names.

Contamination in the Great Lakes

Residues of persistent organics have been found in all of the Great Lakes. Overviews of contamination by organics in the Great Lakes were provided by Delfino (1979) and Kaiser (1979). Several organochlorines were reported in suspended sediments at the mouths of rivers entering the Great Lakes by Frank (1977), although no organophosphorus compounds were observed in the suspended solids fraction.

The occurrence of several organochlorines in Lake Superior sediments were reported by Glooschenko et al. (1976), Veith et al. (1977), Eisenreich et al. (1979) and Frank et al. (1980). Frank et al. (1980)

sampled surficial sediment throughout the lake and found one DDT derivative (p, P'DDE) detectable in 50 percent of the samples, PCB dectable in 44 percent of the samples, and HEOD (Dieldrin) detectable in nine percent of the samples. DDE occurred primarily in depositional basins while PCB concentrations were similar in depositional and non-depositional zones. Eisenreich et al. (1979) reported PCB concentrations of one to two orders of magnitude higher than found in other studies. This can be attributed to the sampling technique of these workers who removed only the surface 0.5 cm layer of sediment for analysis. PCB concentration was found to decline to undetectable levels at four cm. This was interpreted as indicative of the importance of an atmospheric source.

Leland et al. (1973) found that ΣDDT and HEOD were widely distibuted in southern Lake Michigan, Schacht (1974) found elevated levels of ΣDDT and PCB in Illinois sediments. Simmons et al. (1980) found low PCB concentrations in northeastern Lake Michigan in comparison to the other Great Lakes. Murphy and Rzeszutko (1977) concluded that atmospheric deposition is the major source of PCB. Frank et al. (1981) sampled the surficial sediments throughout Lake Michigan and found DDT in 93.4 percent, HEOD in 48.3 percent, HE in 53.9 percent, chlordane in 21.6 percent, and PCB in 79.6 percent of the samples. The distribution of DDT conformed with lake depositional basins following restricted DDT use. The sediment distributions of HEOD, HE, and PCB indicated active use in 1975 and the probable sources. Organochlorine compound levels reported were higher than those for Lake Superior, similar to Lake Huron, and lower than Lakes Erie and Ontario.

Frank et al. (1979 a) measured organochlorine insecticides and PCB's in surficial and core sediments of Lake Huron, Georgian Bay, and the North Channel. DDT residues were present in 92.7 to 97.7 percent of the samples from the three areas and levels of ΣDDT were greater in depositional than in non-depositional zones. All sediments contained detectable concentrations of PCB's and a major source appeared to be Saginaw Bay where Bremer (1978) reported a PCB contamination problem. From the analysis of sediment cores and knowledge of sedimentation rates (kemp and Harper, 1977), Frank et al. (1979 a) determined that ΣDDT and PCB contamination of the lake appeared to begin in the early 1950's. HEOD was identified in 5.7 percent to 30 percent and heptachlor epoxide in 8.5 percent to 23 percent of the samples taken from the three areas. Endosulfan was found in a few Georgian Bay and North Channel samples but none was identified in sediments of Lake Huron proper. No residues of six other organochlorines were detected in any of the samples collected.

Concentrations of organochlorines in Lake St. Clair sediments in 1970 and 1974 were reported by Frank et al. (1977). PCB and ΣDDT were present in all samples in 1970 and in 98 percent and 96 percent of samples taken in 1974, respectively. Mean concentrations of these substances declined significantly over this period due to the implementation of restrictions in their use. HEOD and endosulfan were reported in 24 and 20 percent of the samples respectively, in 1970 but neither were detected in 1974. Five other organochlorines and 20 organophosphorus pesticides were undetected in lake sediments. Heavy PCB contamination of the St. Clair River in the upper industrialized section and subsequent downstream transport were reported by Berg et al. (1974). The distribution of organochlorine contaminants in Lake St. Clair indicated river input to be the primary source of contamination (Frank et al. 1977).

Organochlorine levels in Lake Erie sediments were reported by Frank et al. (1977). All sediments contained detectable ΣDDT and PCB. Concentrations of both substances were highest in sediments of the western basin, reflecting the Detroit River source. Mean PCB and ΣDDT levels found in Lake Erie sediments exceeded those found in the upper Great Lakes. HEOD was present in 86 percent of the sediments analyzed and concentrations also decreased from west to east. While the Detroit River is a major source of contamination in Lake Erie, the Task Force on PCB (1976) estimated that 500 to 1,000 kg PCB/yr enter Lake Erie from atmospheric sources, while input from municipal sewage treatment plants to both Lake Erie and Lake Ontario total approximately 25 kg/yr. Frank et al. (1977) found no detectable chlordane, endosulfan or heptachlor epoxide in Lake Erie sediments, although their detection limits for these analyses were relatively high.

Widespread organochlorine contamination in Lake Ontario sediments was reported by Haile et al. (1975) and Frank et al. (1979b). The latter authors found higher ΣDDT residues in Lake Ontario sediments than reported in other Great Lakes and observed higher concentrations in basin sediments than in non-depositional zones. The distribution pattern of ΣDDT suggested a number of small sources around the lake. PCB's were detected in 96 percent of the samples and mean concentrations were less than observed in Lake Erie but higher than reported in the other Great Lakes. Sediments in depositional zones accumulated higher levels of PCB residues than found elsewhere in the lake. The lake-wide distribution pattern of PCB reflected the importance of the Niagara River as a primary contamination source. HEOD was detected in 39.3 percent of the samples and mean concentrations were less than reported in Lake Erie sediments. Chlordane and endosulfan were detected at a limited number of stations (<10 percent). The distribution and concentration of mirex in Lake Ontario sediments were reported by Holdrinet et al. (1978). Mirex entered the lake via the Oswego and Niagara Rivers and was confined to the surficial sediment of the southern portion of the lake. Jamieson (1977) detected polybrominated biphenyls (PBB's) in industrial wastewaters released into Lake Ontario at Cobourg, Ontario, but residues in lake sediments were not reported.

Sediment-Water Interchange

While trace metals occur naturally in sediments and it is frequently difficult to distinguish between natural and contaminated loads, persistent organics are anthropogenic in origin and their occurrence in the aquatic environment is attributable to pollution.

Many persistent organic contaminants exhibit strong affinities for sediment, while others tend to occur in solution in aquatic environments. Descriptions of studies documenting the sediment-solute distribution of organic contaminants follow.

Organochlorines tend to associate with sediment, while many of the organophosphorus pesticides exhibit relatively high solubilities in water. Studies by Miles (1976) and Miles and Harris (1978) demonstrated that organochlorine insecticides in streams draining agricultural areas in southern Ontario were associated with particulate matter, while some of the organophosphorus pesticides such as diazinon are appreciably soluble in water and were present almost entirely in solution in the same contaminated systems.

Despite the widespread use of organophosphorus pesticides in Ontario, Frank et al. (1977) and Frank (1977) found no detectable organophosphorus residues in Lake St. Clair sediments or in the suspended sediments of several streams flowing into the Great Lakes, respectively.

Berg et al. (1974) observed relatively rapid losses of PCB to sediment from wastewater sources in Ontario. Extremely elevated levels were observed in Hamilton Harbor, particularly near contaminant discharge sources where sediment loads of up to ten ppm and concentrations in water of up to 0.45 ppb were observed. The loss of most PCB residues from wastewater sources to the sediment was also observed in the Ottawa River and the St. Clair River. Greichus et al. (1977, 1978) found that PCB's and organochloride insecticides tended to accumulate in the sediments of African lakes. Hafferty et al. (1977) measured PCB concentrations in water and suspended sediment in the Duwamish River at Seattle, Washington and found a particulate:dissolved PCB ratio of 10^5. Rapid adsorption of PCB by sediment has been reported by several other workers (see Wildish et al. 1980).

The pesticide toxaphene is generally insoluble in aqueous solution and does not leach through soil profiles (Taschenberg et al. 1961; Swoboda et al. 1971; LaFleur et al. 1973). Toxaphene has also been found to accumulate in the sediments of salt marshes (Gallagher et al. 1979).

Oloffs et al. (1978) applied the organochlorine chlordane to a small, uncontaminated lake in British Columbia and traced the movement of pesticide residues in the environment. Concentrations in the water column declined from ten µg/l at the time of application to 4.7 µg/l within one week and to <0.001 ug/l after 421 days. Concomitant increases in sediment levels of chlordane were observed.

Pierce et al. (1977) traced the fate of pentachlorophenol (PCP) in a stream and lake following a spill of this toxic pesticide. Most residues were shown to become sediment-bound. The authors also reviewed laboratory studies on the fate and toxicity of PCP in aquatic environments.

The chlorinated hydrocarbon kepone was associated primarily with suspended sediment during the elutriation of contaminated sediments from the James River which drains into Chesapeake Bay (Saleh et al. 1978).

In a laboratory study, Sikka et al. (1978) demonstrated that >95 percent of the organochloride dichlorobenzidine was adsorbed from solution by lake and pond sediments.

Oloffs et al. (1973) observed that four organochloride pesticides moved into sediments from seawater solution so that no detectable residues remained in solution after six weeks.

Toxic Organic-Sediment Associations

Properties of pesticides that govern their behaviour in the abiotic aquatic environment were summarized by Weber (1977). Four properties are of primary importance: 1) ability to ionize; 2) presence of complexing elements (P, As); 3) solubility in water; and 4) ability to vapourize.

The ability of an organic contaminant to ionize in solution is important in the determination of its behaviour in the environment (Weber 1977). Cationic pesticides (e.g., the herbicides cyperquat, difenzoquet, diquat, paraquat) are quickly adsorbed onto the negatively charged surfaces of suspended sediment and are almost completely removed from solution in the aquatic environment. Anionic pesticides (e.g., the herbicides dalapon, dicamba, picloram, 2,4-D) are only slightly adsorbed by suspended solids and are readily desorbed into solution. Basic pesticides (e.g., the s - triazine herbicides ametryn, atrazine, prometon, prometryn) are readily adsorbed by acidic colloidal surface but are adsorbed to a smaller extent by neutral or basic surfaces. Thus, the adsorption of basic pesticides is pH-dependent. Non-ionic contaminants are adsorbed by colloidal surfaces primarily through physical adsorption forces and retention by particulates depends upon solubility in water and the hydrophilic/lipophilic characteristics of the sediment. Relatively soluble, non-ionic pesticides (e.g., aldicarb, CDAA, dimenoate, molinate, carbetamicle, fenuron, methomyl, propoxur) are adsorbed in small amounts and are readily desorbed from particulates. Organic contaminants of very low water solubility such as aldrin, BHC, lindane, trifluralin, DDT, dieldrin, endrin and oryzalin are adsorbed by highly lipophilic organic sediments but are only slightly adsorbed by hydrophobic clays, and desorb from sediments only slightly. Non-ionic pesticides with complexing groups such as phosphate (demeton, disulfoton, glyphosate, parathion, phorate) and As (DSMA, MAMA, MSMA) are readily complexed and removed from solution by clay colloids.

Weber (1977) also described the influence of contaminant vapour pressure on behaviour in the aquatic environment. Pesticides having high vapour pressures ($>10^5$ mm Hg at 25°C) are considered volatile. Such compounds (e.g., cyperquat, difenzoquat, diquat, paraquat, dalapon, DSMA, glyphosate, MAMA, MSMA, CDAA and the phenoxy acid herbicides) may be adsorbed by particulates but are readily desorbed and subsequently vapourize into the atmosphere. Sediment may compete with the process of volatilization for organics introduced into the aquatic environment. Oloffs and Albright (1974) observed the transport of α-chlordane, γ-chlordane, DDT, PCB (Aroclor 1260) and γ-BHC in water and sediment-water systems. In the absence of sediment, all compounds except γ-BHC were gradually volatilized into the atmosphere. In the presence of sediment, most residues were adsorbed and no volatilization occurred.

Two important sediment properties that influence the adsorption of organic contaminants are sediment particle size and organic matter content. Gallagher et al. (1979) determined that most toxaphene in a Georgia salt marsh was associated with organic constituents of the sediments. Chesters and Lee (1971) investigated the adsorption of a light organochlorine insecticide (γ-BHC) and two organophosphorus pesticides (malathion and parathion) in the laboratory and observed that the most important factors governing adsorption were sediment:water ratio, pesticide concentration, and organic matter content. Richardson and Epstein (1971) showed that the hydrophobic pesticides DDT and methoxychlor tend to concentrate in finer particle sizes while the more soluble endosulfan is preferentially adsorbed by coarser material. Adsorption of the organochlorine herbicide diuron by freshwater sediments was found to be correlated positively with organic carbon content (Peck 1977). Uptake of radio-labelled DDT from seawater by particles ranging in size from bacterial cells to large sand grains was shown to be inversely related to mean particle diameter and to DDT concentration if saturation levels were not exceeded (Hargrave and Phillips 1974). Choi and Chen (1976) found a linear correlation between organochlorine content and both organic carbon content and the fine silt-clay fraction in Los Angeles Harbor sediment. Hiraizumi et al. (1979) experimentally measured PCB adsorption rate onto various particulates. PCB concentration factors of adsorbents were found to be inversely related to particle size and directly related to specific surface area for adsorbents of the same constituent. Concentration factors were also controlled to some extent by organic content, with clays adsorbing much less than muds and plankton. Wildish et al. (1980) observed that a coarse sand with a relatively low organic carbon content became saturated with the PCB Aroclor 1254 when levels of dissolved residue in water reached five μg/l while finer, more organic sediments did not reach equilibria until dissolved concentrations reached at least 40 μg/l. Steen et al. (1978) experimentally demonstrated that both particle size and total organic carbon in freshwater sediments influenced the sediment:water partitioning of four chlorinated biphenyl residues.

Conversely, Stainken and Rollwagen (1979) found no significant correlations between sediment PCB levels and silt-clay content in Raritan Bay, New Jersey. Fulk et al. (1975) found no significant relationship between PCB and organic content in various sediments. PCB concentration in Lake Macatawa, Michigan was influenced more by proximity to contaminant sources than by organic carbon content (Brink 1978). Thus, variations in use patterns within basins and the influence of localized discharge areas may, in some situations, account for the absence of correlations between contaminant concentration and sediment properties.

Studies have also shown that organic contaminant levels in sediment are related to organic carbon and particle size in the Great Lakes. In Lake Huron and Georgian Bay, ΣDDT content was correlated with organic matter content, suggesting sorption or chelation by organic substances (Frank et al. 1979a). While direct relationships of PCB and HEOD with organic carbon were not shown, significant correlations between ΣDDT, HEOD and PCB concentrations reflected the general association of these compounds. Glooschenko et al. (1976) similarly found no significant correlation between organic carbon and PCB concentration in the sediments of Lake Huron and Lake Superior. Simmons et al. (1980) and Leland et al. (1973) found linear relationships between organochlorine concentrations and both the fine silt-clay fraction and organic carbon content in Lake Michigan. In Lake Erie, DDE, TDE and PCB showed good correlations with organic carbon content, but HEOD concentrations were unrelated to other sediment parameters (Frank et al. 1977). It was concluded that relationships observed between Hg, ΣDDT and PCB's reflected not only their sediment associations, but also their similarity of source and dispersion in Lake Erie. In Lake Ontario, Frank et al. (1979b) demonstrated that ΣDDT components were highly interrelated and each was significantly correlated with organic carbon, the clay size fraction and mean grain size. PCB's showed similar but poorer correlations with the same parameters. Again, the lack of good correlations between some organochlorine levels and other sediment properties in certain instances likely reflect variations in basin use patterns and in the influence of localized contaminant sources, as suggested by Frank et al. (1979a).

Lambert (1966-1968) suggested that organic matter acts similarly to an organic solvent and that the partitioning of a neutral organic compound between soil organic matter and water should correlate well with its partitioning between water and an immiscible organic solvent. Briggs (1973) demonstrated a correlation between soil sorption of phenyl urea herbicides and their octanol-water partitioning. Karickhoff (1979) investigated the partitioning of several aromatic and chlorinated hydrocarbons between various sediments and water and demonstrated that octanol-water distribution coefficients can be used to estimate partition coefficients between water and sediment organic matter. Adsorption was found to be directly related to organic carbon content for a given particle size isolate.

Sediment-solute distributions of organic contaminants tend to reflect the solubilities of these compounds in water. As mentioned earlier, organochlorine compounds exhibit relatively low solubilities in water and, consequently, tend to associate with sediment. For instance, the solubilities of DDT, dieldrin and PCB are 3.4, 12.5 and 100 to 1,000 µg/lc, respectively (see Kent and Johnson 1979) and these compounds exhibit strong affinities for particulate matter. Organophosphorus compounds, however, are often appreciably soluble in water and may be almost entirely present in solution in contaminated systems (Miles 1976). Miles (1976) observed that concentrations of DDT, TDE, dieldrin and diazinon in suspended stream sediments in Ontario were consistent with their solubilities. While PCB's exhibit low solubilities and are preferentially adsorbed onto solids in general (Crump-Wiesner et al. 1973; Hammon et al. 1972), solubility decreases and adsorption increases with degree of chlorination (Haque and Schmedding 1976). Halter and Johnson (1977) demonstrated that four chlorine isomers have greater water solubilities than more highly chlorinated biphenyl compounds. Relative solubilities and adsorptivities of PCB residues were also studied by Steen et al. (1978). Solubilities of 738 pesticides in water were reported by Gunther et al. (1968).

Once adsorbed, organochlorine residues do not readily desorb (Huang et al. 1974; Wildish et al. 1980). Huang (1974) speculated that strong ionic interactions and hydrogen bonding stabilize organochlorine-sediment interaction. While most workers agree that adsorption of PCB's by sediments occurs rapidly (e.g., Muto et al. 1974; Hiraizumi et al. 1979; Steen et al. 1978; Wildish et al. 1980), reported rates of desorption range from rapid (Steen et al. 1979) to very slow and negligible (Haque et al. 1974; Hiraizumie et al. 1979). Rapid desorption of more soluble, four chlorine residues (tetrachlorobiphenyl) and very slow desorption of more highly chlorinated, less soluble PCB residues were reported by Halter and Johnson (1977).

The study of Berg et al. (1974) demonstrated that hydrologic characteristics of aquatic systems influence organochlorine sedimentation. PCB's were quickly lost to the sediment near the point of discharge into the static system of Hamilton Harbour and into a slow-flowing Ottawa River impoundment. Conversely, the dynamic system of the St. Clair River favoured downstream transport of PCB's. Static and flowing conditions have also been found to influence sediment-solute distributions of PCB's (Halter and Johnson 1977).

Few studies have been carried out on the effects of other environmental factors such as temperature, dissolved oxygen, pH, and salinity on the interaction of organic contaminants with aquatic sediments. Peck (1977) demonstrated that adsorption of the organochlorine diuron was inversely related to temperature. Huang et al. (1971) studied the adsorption and desorption of DDT, dieldrin and heptachlor by clay minerals under conditions of varying temperature, pH, salt content, and dissolved organic content. None of these factors appeared to exert an influence on sediment-contaminant behaviour. Anaerobic conditions may promote the microbial decomposition of some organic compounds. For instance, the reductive dechlorination of DDT to the less toxic compound TDE is accomplished by anaerobic bacteria (Miles 1977). While the adsorption of basic pesticides such as the s-trizine herbicides is pH-dependent (Weber 1977), apparently no studies have been conducted to investigate the influence of pH on organic contaminant behaviour in aquatic systems.

Huang et al. (1974) investigated the behaviour of three chlorinated hydrocarbons - DDT, dieldrin and heptachlor - in experimental systems containing various clay minerals and organic matter and described sorption-desorption phenomena on a molecular basis. The adsorption and desorption of pesticides to and from three types of clay, (montmorillonite, illite and kaolinite), each exhibiting a distinct chemical structure and character, and an organic-rich sediment were studied. Adsorption on non-expansible clays was nearly instantaneous since pesticides can only be deposited on the surfaces and edges of clay particles. Adsorption onto the expansible clay montmorillonite generally occurs in two stages: the pesticide is instantaneously deposited on the clay surface, and then additional pesticide diffuses into the clay's interlamellar spaces. Dieldrin, however, did not follow this two-stage adsorption pattern due to the physical inability of dieldrin molecules to diffuse into these spaces. Organic-rich substances adsorbed pesticides more slowly than clays but had much higher sorptive capacities. The lower rate of sorption on humic substances was attributed to the presence of a hydrophilic barrier surrounding the organic particles. The higher sorptive capacity of humic substances was attributed to the presence of a great many functional groups on humic molecules which form dipole-dipole, ion-dipole or hydrogen bonding interactions. Adsorption in all cases was relatively irreversible.

Many toxic organic pollutants may be degraded by the process of photolysis. For example, Sikka et al. (1978) demonstrated that dichlorobenzidine (DCP) in aqueous solution has a half-life of only about 90 seconds when exposed to sunlight or to artificial light in the laboratory. While many toxic organics may be susceptible to photolysis, contaminant associations with sediments and organic matter may inhibit photodegradation (Hamadmad 1967; Crosby 1972; Pierce et al. 1977). Bunce and Chittim (1979) pointed out the difficulty in determining whether solar photodegradation is of any importance under natural conditions. Processes of chemical reactions, biotransformations and translocation all compete with photolysis in nature.

Whitmore (1977) modelled the sources, distribution, and fate of PCB's in the environment and applied the model to the Great Lakes. Atmospheric sources were determined to be a major source of PCB contamination in the lakes. This is in accordance with the Task Force on PCB (1976), Murphy and Rzeszutko (1977) and Eisenreich et al. (1979) who concluded that atmospheric deposition is a major PCB source in the Great Lakes.

Lassiter et al. (1979) developed a model describing the fate of toxic organic substances in aquatic environments. The model considers environmental and chemical variables known to influence the behaviour of these compounds.

EFFECTS OF BIOTA ON SEDIMENT-BOUND METALS

This section reviews the literature concerning the effects of various components of biological communities on the speciation, concentration, and distribution of metals in sediments. While biogenically-derived organic matter such as humic acids can be extremely important in the biogeochemical cycling of trace metals, this represents an indirect effect of biota and has been considered previously (Sediment Contamination by Trace Elements).

Microbes

Microorganisms exert a considerable influence on trace metals in aquatic ecosystems. Kusnetsov (1975) demonstrated that microorganisms are partially responsible for the decomposition of higher molecular weight organic acids which might be associated with metals to smaller, more soluble forms. Microorganisms also mobilize metals from sediments by specific biochemical means. Ramamoorthy et al. (1977) found that bacteria compete effectively with sediment in accumulating mercuric ions from water and postulated that bacteria should have a significant effect on the mobilization of Hg from sediment into food chains. Bacteria are also known to bioconcentrate Am-241 and may play an important role in the mobilization of this metal from sediment (Giesy and Paine 1977).

The importance of the biological methylation of heavy metals is well documented. Methylation increases toxicity and potential for bioaccumulation of heavy metals and plays an important role in sediment-water interchange (Chau and Wong 1978). Although it is a widely held hypothesis that microbial methylation of Hg in sediments is responsible for methylmercury production, the precise role of microorganisms in mercury methylation is not fully understood.

Methylmercury is highly neurotoxic and may be the most important species involved in the mobilization of Hg from sediments to biota. Information on the relative significance of methylmercury in contaminated fish is conflicting, although most data indicate that the mercury burden in fish is predominantly in the form of methylmercury (see Bishop and Neary 1974; DeFreitas et al. 1974). Conversely, the proportion of methylmercury in sediment is much lower (Kudo et al. 1977b). In fact, while methylation undoubtedly occurs in some sediments, Spangler et al. (1973) pointed out that methylmercury may be undetectible in sediments where Hg contamination is heavy.

The biological production of methylmercury and dimethylmercury in sediment was demonstrated by Jensen and Jernelov (1969). Bacteriological methylation of mercury was studied by Wood et al. (1968) and Imura et al.(1971). Methylation of Hg by pure curltures of fungi (Landner 1971; Vonk and Sijpesteijn 1973) and bacteria (Tonomur et al. 1972; Vonk and Sijpesteijn 1973; Blair et al. 1974; Huey et al. 1974) have been demonstrated. The abiotic methylation of Hg by biologically methylated Sn was observed by Huey et al. (1974) who suggested that this may be a pathway for Hg methylation in environments where these metals are co-contaminants. Kushner (1974) was unable to demonstrate any biological methylation of Hg in Ottawa River sediment. The apparent absence of methylmercury in some Hg-contaminated waters (Spangler et al. 1973) may be attributable to the rapid conversion of sediment-bound methylmerucry to dialkylmercury and possibly to mercuric forms by bacteria (Sommers and Floyd 1974).

Saitoh and Cheam (1975) showed considerable variation in methylmercury levels of sediments from Lake St. Clair, Port Stanley (Lake Erie), the Niagara River mouth, and Hamilton Harbour. Methylmercury concentrations were independent of total Hg levels. The upper limit of methylmercury content expressed as a percentage of total Hg approached one percent for Lake St. Clair sediments.

The methylation of lead by microorganisms in lake sediments was first demonstrated by Wong et al. (1975). Methyllead, like methylmercury, is highly toxic and is readily accumulated by aquatic organisms (Wong et al. 1975, Wong and Chau 1979). Sediments from Lakes Ontario, Erie, and St. Clair were incubated by Wong et al. (1975) who found that inorganic Pb was converted to methyllead by purely biological processes.

The microbiological methylation and transmethylation of As (Wood 1973) and Se (Chau et al. 1976, Chau and Wong 1978) have also been described.

Primary Producers

Algae and aquatic vascular plants bioconcentrate trace elements, and thus mediate in the transport of metallic contaminants between sediment and water.

Mechanisms by which aquatic vegetation binds heavy metals have been instigated by Gale and Wixson (1979). Some particles containing heavy metals are physically trapped by plants. Heavy metal ions tend to displace normal cations such as Na^+, K^+, Mg^{2+} which are commonly associated with anionic sites on and within cell walls. Cationic exchange capacities of filamentous algae for lead were also determined. Wahlgren and Nelson (1974) estimated the half-lives of Pu-237 and Cs-137 in Lake Michigan and demonstrated that sorption by phytoplankton and the subsequent rapid settling of phytodetritus and zooplankton fecal pellets were likely responsible for rapid sedimentation of Pu-239. Investigations on the feasibility of removing trace metals from wastewaters by algae have been undertaken. Jackson (1977, 1978) found that green algae generated by sewage led to the entrapment of metals from mining wastes into sediments of Schist Lake, Manitoba. Algal seston was found to preferentially bind Zn and Cu. The subsequent production of hydrogen sulphide by the decomposition of organic waste and algae resulted in the formation of stable metal sulphides which accumulated in lake muds.

A process for the biological treatment of mining and milling wastes containing high levels of heavy metals was described by Gale and Wixson (1979) and Jennett et al. (1979). The "algal meander system" was developed when it was observed that lagoon effluents from Pb/Zn mining and milling operations were inducing heavy growth of algae in receiving streams. This system uses a series of meandering channels from the lagoon discharge to the receiving water. Algae in the meanders were shown to be 99+ percent efficient in heavy metal removal from the wastewater. Experiments conducted by Jennett et al. (1979) demonstrated that the efficiencies of several species of bluegreen algae, green algae, and diatoms in the removal of As, Cd, Pb, Hg, and Zn from solution varied interspecifically. As algae die and settle to the bottom, the heavy metals sorbed by the plants are incorporated into the sediment. Algae also play a role in the mobilization of metals from sediment to water. Ankistrodesmus braunii and Anabaena sp. accumulated heavy metals from contaminated sediment in experiments carried out by Laube et al. (1979), although sediment concentrations of metals did not decrease appreciably. Giesy and Paine (1977) observed the uptake of Am-241 by Scenedesmus and Selenastrum and concluded that algae may participate in the mobilization of this element from sediment in natural aquatic environments.

Other studies have shown that rooted aquatic plants may play a role in the transport of metals into sediments. In laboratory experiments on Ottawa River sediments, Mortimer and Kudo (1974) observed that sediment in which Elodea had grown contained twice as much Hg as plant-free sediment. The same workers (1977) fround no correlation between Hg in sediment and absorption by macrophytes and found no indication that roots mediate in the release of Hg from sediment. Furthermore, the laboratory experiments of Mortimer (1977) demonstrated that aquatic plants from the Ottawa River accumualte Hg from water while discriminating against uptake from sediments.

Rooted plants may also mediate in the flux of trace metals from sediment. Welsh and Denny (1976) proposed that Pb is absorbed from sediments through the roots of aquatic plants and is subsequently released into the water from decaying leaves. Mayes et al. (1977) demonstrated in field experiments that concentrations of Pb and Cd differed significantly in Elodea canadensis grown in the same water but in sediments with different levels of the two metals, indicating that there was removal of Cd and Pb from the substrate. Chittenden (1976) observed lower levels of Mn and Fe in weed bed sediments than in deeper areas, suggesting a possible biological mediation in removal from sediment. In the Moira River, a tributary of Lake Ontario, concentrations of Ni, Co, and Cu in bottom sediment were correlated with concentrations in Myriophyllum verticillatum and Elodea canadensis, suggesting biological uptake from the substrate (Mudroch and Capobianco 1979).

Kozuchowski and Johnson (1978) raised the interesting prospect that emergent macrophytes may remove Hg from sediment by direct injection into the atmosphere. The authors correlated sediment Hg levels in a New York lake with concentrations released by the reed Phragmites communis into the atmosphere. The diurnal patterns of Hg release suggested a connection with photosynthesis or transpiration.

The influence of aquatic macrophytes on metals in sediments is dependent upon physiocochemical conditions. Patrick (1977) demonstrated that uptake of Cd from sediments by marsh plants was more related to sediment conditions than were other metals, and that the rate of uptake increased with Eh. Zinc uptake was also directly related to Eh. Mercury accumulation from sediment increased with increasing pH and with declining salinity. Uptake of Cu from sediment was little affected by Eh or PH.

Aquatic macrophytes can also influence sediment chemistry by altering pH, Eh, sulphide levels, and sediment organic levels and composition, not only through decomposition after death, but also during active growing stages. For instance, Tessenow and Baynes (1975) demonstrated that an oxidized layer in surficial sediments was maintained by oxygen transport through the tissues of the hydrophyte, Isoetes lacustris, resulting in accumulations of Fe and Mn oxides at the oxidation-reduction interface.

Benthic Invertebrates

Through the process of bioturbation, sediment-dwelling benthic organisms can influence metal distribution and physiochemistry in the sediment.

Tubificids have been considered the most important mixers of surface sediment in freshwaters (Davis 1974). Prosi (1979) observed the effects of tubificids on sediment metal distribution in aquaria. Worms caused an enrichment of metals in surface sediment due to the ingestion of deeper contaminated sediment and the egestion of contaminated fecal matter near the sediment-water interface. The metals in this fecal matter were then gradually released to the overlying water. Boddington et al. (1979) concluded that benthic macrofauna may be entirely responsible for the clearance of Hg from sediments in flowing waters. These workers investigated the influence of Hg in Ottawa River sediments in the laboratory and found that the clearance rate of inoranic Hg and methylmercury from sediments was linearly related to worm density. The most significant loss was from the top centimetre of sediment.

Krezoski et al. (1978) investigated radioactivity profiles in Lake Huron sediment and demonstrated an agreement between the depth of the surface layer of homogeneous radioactivity and the vertical distribution of benthic macroinvertebrates (primarily Oligochaeta and Amphipoda). The authors considered oligochaete defecation rates, amphipod burrowing depths, and benthic densities and concluded that bioturbation of sediments by zoobenthos is probably important in the mixing of profundal sediments in the Great Lakes.

Fish

Fish may also influence the behaviour of trace metals in sediments. Kudo and Mortimer (1979) found that the amount of Hg accumulated by fish in aquaria is nine times greater than the amount accumulated from water in which Hg had come from bed sediments. Koyanagi et al. (1978) also demonstrated direct biomobilization of trace metals from sediment by fish. While such studies are primarily concerned with the significance of bioaccumulation to the biological community, they also indicate that activities of fish exert an influence on metals in sediments.

Man

While the obvious influence of man on trace metals in sediments is the anthropogenic loading of metal-contaminated wastes to natural waterways, this is a vast topic in itself and is beyond the scope of this review. However, effects of human activities on insitu metals in sediments are relevant.

The mechanical disturbance of sediments can exert major influences on the concentrations and distributions of metals in aquatic sediments. Major processes of concern here are dredging and dredged spoils disposal. Dredging activities affect sediment quality by re-distributing and transporting contaminated sediment from areas such as harbours and shipping channels to less contaminated disposal sites and by re-mobilizing sediment-bound heavy metals.

The presence of heavy metals in dredged sediments prompted the development of dredged material disposal criteria designed to minimize adverse environmental impacts of contaminated sediments on water quality. The bulk chemical content of sediments should not be used in the regulatory process since the amounts of heavy metals available to become dissolved into the aqueous phase are unrelated to the total content of metals in sediments (Lee et al. 1975).

The "standard elutriate test" was included by the United States Environmental Protection Agency (USEPA) (1975) in a set of guidelines for the evaluation of the pollution effects of dredged spoils disposal. The procedure is to combine tests sediment to water from the proposed disposal site in a 1:4 volume ratio of sediment:water. The mixture is vigorously shaken for 30 minutes, allowed to settle for one hour, and filtered or centrifuged. Elutriate testing may be carried out under varying physiocochemical conditions appropriate to conditions in the receiving water (e.g., Lee et al. 1978). Appropriate testing is then carried out on the elutriate to predict the release of contaminants from sediment to water during disposal. Procedures for elutriate testing have been detailed elsewhere (Keely and Engler 1974; Army Engineer Waterways Experiment Station 1976; USEPA/U.S. Corps. of Engineerings 1977).

While elutriate testing is superior to bulk chemical analyses of sediments, the procedure is not without shortcomings. Cheam et al. (1976) prepared elutriates of Hamilton Harbour (Lake Ontario) sediments and found that complexing organic agents extracted by elutriation of sediments were in higher concentrations in elutriates than in disposal waters. This resulted in excessive complexing of heavy metals in elutriates in comparison with dredged spoil receiving waters. Grimwood and McGhee (1979) compared observed pollution effects with effects predicted by the standard elutriate test using several sediments from the lower Mississippi River basin. Analysis of effluent and elutriate samples indicated that for Cr, Cd, and, to a lesser extent, As, Ni, and Zn, the elutriate test offered a useful prediction of pollution from dredging. However, the test failed to offer a reliable prediction of release of Pb, Hg, and Cu due to a negative release (i.e., adsorption) of these metals by suspended solids during disposal.

Physiochemical conditions at disposal sites largely determine the extent of trace metal release during dredging. Dredging very frequently involves the mixing of reduced sediments with an oxidizing aqueous environment. Gambrell et al. (1977) and Khalid et al. (1979) demonstrated that changes in pH and Eh during dredging activities regulate the chemical availability of potentially toxic metals. Once oxygenation of the sediments begins, sulphide is rapidly oxidized to elemental sulphur and then further oxidized biologically to sluphuric acid. The reduced and relatively soluble forms of Fe and Mn are oxidized to the insoluble ferric and manganic forms. Conversely, the reverse processes may occur when oxygenated fill material is deposited into anaerobic waters. As discussed previously, these processes have pronounced effects on trace metal chemistry. Patrick et al. (1977) provided an extensive discussion of physiocochemical factors regulating heavy metals in dredged materials. A literature review by Khalid et al. (1977) stressed the effects of pH and redox potential on sediment metal chemistry. They suggested that some site-specific environmental impact studies be carried out for each proposed dredging project due to the myriad of potential interactions of dredging activities with the environment.

Sly (1977) summarized research on the effects of dredging in Canadian waters of the Great Lakes on trace metal chemistry. Studies on pore water concentrations of selected elements in recently dumped sediments forming an island in Lake St. Clair were undertaken by Mudroch and Cheam (1974). Sediment concentrations of eight heavy metals showed similar profiles with depth. Pore water concentrations of heavy metals were largely unrelated to sediment composition. Data reported by Chemex Laboratories Limited (1974) demonstrated that levels of heavy metals, with the exception of Fe, in water columns at Port Stanley Harbour (Lake Erie) and Bronte Harbour (Lake Ontario) changed little during dredging and water quality met Canadian drinking Water standards.

Contaminant release from bioturbation and from wave action and storm seiches appears to be the most important source of internal loading by far in the Great Lakes (Sly 1977). Chemex Laboratories Limited (1973), using turbidity measurements, estimated that under the conditions prevalent in November during their study, at least 1,000 metric tons of suspended solids were naturally present in Lake Ontario for each kilometre of shoreline near Bronte Harbour. This natural loading was many times greater than loadings due to the dumping of dredged materials. Kemp et al. (1976) estimated that dredging and dumping only accounts for about four percent of the annual lake loading of silt and clay in Lake Erie. Although the concentrations of trace metals are generally higher in dredged materials than in most sediment, the data presented in these studies help place the significance of these sediment loadings into perspective.

EFFECTS OF BIOTA ON SEDIMENT-BOUND ORGANICS

This section reviews the literature on the effects of various components of biological communities on the speciation, concentration, and distribution of toxic organics in sediments. While biogenically-derived organic matter exerts an important influence on the partioning of toxic organic contaminants, this represents an indirect effect of biota and has been considered elsewhere (Sediment Contamination by Persistent Organics).

Microbes

Microbes influence the transport of organics from solution and suspension to the bottom sediment. The association of chlorinated hydrocarbons with suspended microparticulates in Lake Erie was investigated by Leshnioswky et al. (1970) and Pfister et al. (1971) who demonstrated that floc-forming bacteria isolated from Lake Erie concentrate the pesticide aldrin associated with suspended colloids and subsequently settle from the water column. The former workers also showed that aldrin is concentrated 625 times over dissolved concentrations in Lake Erie water by these floc-forming bacteria.

While the studies of Pfister et al. (1971) and Leshnioswky et al. (1970) inidcated that bacteria mediate in the formation of sediment-contaminant associations, Hargrave and Phillips (1974) found that the presence or absence of microorganisms did not influence the adsorption of DDT by various particulates in short-term laboratory experiments.

Many investigations have demonstrated that microbial metabolism may chemically transform toxic organics. DDT and BHC degrade rapidly under anaerobic conditions, apparently due to reductive dechlorination by anaerobic bacteria (Guenzi and Beard 1967; Lotse et al. 1968; Miles 1977). Oloffs and Albright (1974) observed the microbial degradation of organochlorines in sediment-water systems in the laboratory. A 75 percent loss of γ-BHC in an unautoclaved system after three weeks could be ascribed to microbial activity. The slight conversion of DDT to TDE (DDD) was also attributed to microbial action. DDT metabolites may predominate over concentrations of the parent compound in sediments (Frank et al. 1974). The relative importance of DDT and its metabolites in Great

Lakes sediments were reported by Frank et al. (1977; 1979; 1979b, 1980) and Haile et al. (1975). Furukawa and Matsumura (1975) isolated a bacterial strain belonging to the genus Alkaligenes from a Wisconsin lake sediment using PCB as a sole carbon source. These bacteria metabolized various PCB residues, including a highly chlorinated form, through an oxidative pathway. Degradation increased as the degree of chlorination decreased. PCB's were oxidized in a two-step process to chlorobenzoic acids. Halter and Johnson (1977) also observed that more soluble, less chlorinated PCB residues are more susceptible to microbial degradation. The experiments of Kempe (1973) demonstrated that the lampricide TFM is degraded microbiologically in sediment-water systems and concluded that microbial action also accounts for degradation in natural streams. Oloffs et al. (1978) measured the disappearance of chlordane from an artificially contaminated lake and discussed the loss of persistent constituents in terms of microbial metabolism and chemical reaction. Hamadmad (1967), Crosby (1972) and Pierce et al. (1977) suggested that toxic organics may persist in sediments due to their association with the organic matter which may inhibit microbial degradation.

Primary Producers

Phytoplankton may act like bacteria in adsorbing toxic organic compounds from water, thereby mediating in transport to the sediment. Hiraizumi et al. (1979) demonstrated that phytoplankton cells are relatively efficient in the adsorption of PCB from solution largely due to their high organic content. Zarull (1975) concluded that DDT is adsorbed onto the lipid fraction of the trilaminer external cell wall component of the chlorophyte Scenedesmus quadricanda. Contaminants may then be transported to the sediment on dead algal cells or in zooplankton fecal pellets.

Aquatic macrophytes may also influence the distribution of toxic organics in aquatic systems. Gallagher et al. (1979) demonstrated that the hydrophobic pesticide toxaphene associates with the roots of the saltmarsh hydrophyte, Spartina alterniflora, and was subsequently removed from sediment by transport to the shoots. Also, dead plant roots adsorbed considerably more toxaphene than did other sediment constituents.

Invertebrates

Zooplankton, like phytoplankton, may also mediate in the transfer of organic contaminants from water to sediment. Hiraizami et al. (1979) showed that on a per unit surface area basis, zooplankton adsorbs PCB more efficiently than any of of the other particulate substances investigated. Also, bioaccumulation of contaminants by zooplankton through the consumption of contaminated phytoplankton may also occur. Upon death, zooplankton and associated contaminants settle and become incorporated into the sediment.

Benthic invertebrates may influence contaminant distribution and physicochemistry in sediment. Polikarpov et al. (1979) and Elder et al. (1979) demonstrated that marine organisms mobilize and accumulate PCB from contaminated sediment. Freshwater benthos accumulate chlorinated hydrocarbons in areas of high sediment contaminant levels (Greichus et al. 1977, 1978; Sarkka et al. 1978). While such studies are concerned primarily with bioaccumulation, uptake of contaminants by benthos may regulate sediment contaminants to some degree. Benthic organisms are important mixers of freshwater sediment (Davis 1974; Krezoski et al. 1978; Boddington et al. 1979; Prosi 1979) and may alter toxic organic distribution both directly through burrowing and bioaccumulation and indirectly by altering sediment properties and, therefore, adsorptive characteristics. Apparently, studies on the role of benthic bioturbation on toxic organic physicochemistry in sediment have not been carried out.

Fish

Fish may remove and bioaccumulate toxic organic compounds from sediment, thereby altering contaminant concentration and distribution. Halter and Johnson (1977) found that fathead minnows (Pimephales promelas), allowed direct access to PCB-contaminated sediments, accumulated PCB at six times the rate of those screened from the sediments and suggested that bioturbation of contaminated sediments results in greater desorption and subsequent bioaccumulation. The experiment of Kobylinski and Livingston (1975) suggested that the hogchoker (Trinectes maculatus) may accumulate significant amounts of mirex from contaminated sediments. The presence of high tissue concentrations of organochlorines in fish of the Great Lakes basin (Reinert 1970; Reinke et al. 1972; Berg et al. 1974; Schacht 1974; Frank et al. 1974; 1978; Haile et al. 1975; Delfino 1979; Kaiser 1979) and in other aquatic systems (e.g. Greichus et al. 1977, 1978; Young et al. 1977; Kent and Johnson 1979) indicate that fish effectively accumulate toxic organic substances from the environment. While these studies are concerned with the significance of bioaccumulation to the biological community, they also suggest that fish exert an influence on contaminants in the physical environment.

Man

The obvious influence of man on toxic organic substances in sediments is the manufacture of these materials and subsequent discharge directly (in wastewaters) and indirectly (through runoff and atmospheric release) into the aquatic environment. While this vast topic is beyond the scope of this review, the effects of human activities on in situ metals in aquatic sediments are relevant.

The mechanical disturbance of sediments influences the concentration and distribution of associated toxic organics. Dredging activities affect sediment quality by re-distributing and transporting contaminated sediment from areas such as harbours and shipping channels to less contaminated disposal sites and by re-mobilizing organic contaminants into the water column.

Dredged material disposal criteria developed by the U.S. Environmental Protection Agency were designed to minimize adverse environmental impacts on water quality. Procedures for conducting elutriate testing to determine contaminant release potential of sediments during mechanical disturbance are described elsewhere in this review (Effects of Biota on Sediment-Bound Models).

Dredging frequently involves the mixing of reduced sediment with an oxidized aqueous environment resulting in changes in pH and Eh conditions. Changes in physicochemical conditions during dredging would probably influence microbial community composition. In turn, the microbial degradation of toxic organics would be affected, although apparently no studies have addressed this matter.

Few studies have addressed the problem of toxic organic release due to the mechanical disturbance of sediment. Dredging of PCB-contaminated sediment in the Duwamish River estuary at Seattle did not result in a significant increase in dissolved or adsorbed PCB concentration in the water column (Hafferty et al. 1977). Results of elutriate tests as well as short-term and long-term expeiments performed to simulate dredging operations indicated insignificant release of PCB from contaminated Los Angeles Harbour sediment (Chen and Wang 1976). Elutriates of contaminated Hamilton Harbour, Lake Ontario sediments did not contain higher concentrations of DDT or PCB than observed in the water column of the harbour (Cheam et al. 1976). Murakami and Takeish (1977) demonstrated in the laboratory that significant amounts of dissolved PCB were not released from contaminated Japanese sediments when mixed with water. Conversely, Lee et al. (1978) described significant releases of various organochlorine residues during elutriation of several contaminated sediments from the U.S. Contaminant release varied both with organochlorine residues and with sediments.

The disturbance of bottom sediment by ship passage is important in shallow harbours and channels and the effect of shipping in sediment disturbance likely exceeds the effects of dredging and disposal by at least an order of magnitude in the Great Lakes (Sly 1977). Thus, Great Lakes ship traffic likely contributes significantly to the resuspension of sediments contaminated with toxic organic compounds.

The effects of wave action and storm seiches in the disturbance of sediment likely far outweigh the effects of dreding and shipping, as discussed in an earlier section (Effects of Biota on Sediment-Bound Metals).

IMPACT OF METAL-CONTAMINATED SEDIMENT ON BIOTA

Biological Uptake

In addition to the hazard of direct toxicity to organisms, biological uptake of trace metals may lead to modification of food webs and toxicity to man through the consumption of contaminated seafoods.

In this review, "bioaccumulation" and "biomagnification" are used as defined by Isensee et al. (1973). Bioaccumulation is used to indicate how extensively an organism accumulates a substance from its surrounding environment by all processes. Biomagnification concerns the process of concentration through the consumption of lower trophic level organisms by higher trophic level organisms with a net increase in tissue concentrations. "Bioconcentration" is used to designate bioaccumulation where tissue concentrations of contaminants exceed concentrations in the external environment.

The biological uptake of trace metals by aquatic organisms is well documented (e.g., review by Leland et al. 1979). While numerous studies have shown that sediments are sinks for most trace metals (Thomas 1972, 1983; Kemp et al. 1978), the relative contribution of contaminated sediment in the process of bioaccumulation has been studied less extensively.

Bioaccumulation

Microbes

The importance of bacteria in trace metal accumulation by sediment and in mobilization from sediment is well known (Ramamoorthy et al. 1977). Bacteria can increase the availability of trace metals to other organisms indirectly by mediating the conversion of metallic contaminants to more biologically available forms and directly by accumulating metals from the environment and serving as food sources to bacteriophagous organisms. This latter process was documented by Colwell et al. (1975)

who found that bacteria labelled with Hg-203 are taken up by bactiverous ciliates and by oysters, resulting in Hg bioaccumulation in the predators.

Phytoplankton and aquatic macrophytes

Algae and aquatic macrophytes accumulate trace metal contaminants from sediment. Laube et al. (1979) observed the accumulation of Cd and Cu from sediment by the freshwater alga Anabaena (Cyanophyta) and Ankistrodesmus (Chlorophyta) but concluded that water was the medium of transport. Lead concentrations in micro-algae and in the aquatic macrophyte Vallisneria grown in an experimental system of water and Pb-contaminated sediment were compared with Pb concentrations in a relatively uncontaminated system (Milne and Dickman 1977). While four times more Pb was desorbed from contaminated sediments than from control sediments, Pb levels in the algae were 10 to 30 times greater and levels in Vallisneria were two times greater than in the corresponding plants in control systems. Elevated levels of several elements have been found in plants growing in watersheds containing coal ash in the sediments (Rodgers et al. 1978). Gentner (1977) observed the uptake of Fe by Vallisneria and demonstrated that uptake from sediments by roots exceeded uptake from water by shoots. Several other studies have demonstrated biological uptake of metals from sediments by aquatic plants (Chittenden 1976; Mayes et al. 1977; Patrick 1977; Mudroch and Capobianco 1979), although studies by Mortimer (1977) demonstrated that some vascular plants may accumulate metals from water while discriminating against uptake from sediment. Similarly, the observations of Ueda et al. (1978) on marine algae showed that the influence of sediment is considerably less than the influence of seawater on the bioaccumulation of Ru-106/Rh-106 and Cs-137.

Benthic Invertebrates

Benthic invertebrates are known to accumulate trace metals in areas of sediment contamination. Eyres and Pugh-Thomas (1978) demonstrated correlations of Pb in invertebrate tissue with substrate concentrations in an English river, although tissue Cu levels correlated poorly and tissue Zn levels did not correlate with substrate concentrations of these metals. Vermeer (1972) and Sheffy (1978) observed high levels of Hg in crayfish in watersheds containing Hg-contaminated sediment in Canada and Wisconsin, respectively. Bissonnette (1977) demonstrated considerable uptake of Hg and Pb by chironomids over sediment concentrations in Washington lakes.

Several studies of metal uptake from sediments by marine benthos have been undertaken. Bryan and Uysal (1978) concluded that ingestion of sediment was the primary mode of uptake of Pb, Cd, Co, Cr, Ni and Zn while dissolved forms of Fe and Mn contributed most to accumulation of these metals in the clam, Scrobicularia plana in an English estuary. Luoma and Jenne (1975) and Neff et al. (1978) also observed bioaccumulation of sediment-bound heavy metals by deposit-feeders. The polychaete Nereis virens accumulates Cd from sediment, although uptake was from readily dissociated forms and the rate of uptake was equal to the rate of dissolution from sediment (Ray et al. 1980). Schmidt (1978) noted that polychaetes tend to directly adsorb Cu from sediment interstitial water.

Fish

Fish may also accumulate trace metals directly from sediment. Delisle et al. (1975) studied bioaccumulation of Zn, Pb, Cu and Cd from sediment by fish and noted that foraging habits and proximity to the sediments appeared to exert a significant influence on the degree of uptake by different species. Gillespie and Scott (1971) and Gillespie (1972) observed the uptake of Hg from various sediments by guppies (Poecilia resticulata). Koyanagi et al. (1978) determined that demersal fish may bioaccumulate trace metals by incidentally ingesting contaminated sediment during feeding. Kudo and Mortimer (1979) observed the accumulation of Hg by fish from sediment and water and demonstrated that nine times more Hg was accumulated from sediment than from water.

Traversy et al. (1975) measured As and Se in water, sediments, and fish from Lakes Erie and Ontario and found the highest concentrations in sediment and the lowest concentrations in water. The relative importance of sediment and water in bioaccumulation by fish was not investigated.

Biomagnification

Metals may show step-wise increases in tissue concentration with increasing trophic level due to food chain biomagnification. Biomagnification of Hg from sediment to bacteria to ciliates and from bacteria to oysters was demonstrated by Colwell et al. (1975). Food chain biomagnification of Hg was also observed by Sarkka et al. (1978) in a Finnish lake with Hg-contaminated sediment.

In studies involving the biological uptake of several elements in contaminated systems, the behaviour of metals in food webs has been found to vary considerably. Cherry and Guthrie (1977) and Guthrie and Cherry (1979) examined the concentrations of ten toxic elements in the water, sediment, aquatic plants, invertebrates, and vertebrates of a coal ash basin and its drainage system in South Carolina and demonstrated that biota bioaccumulate various toxic metals at different rates. Copper was apparently biomagnified in the food chain with the highest concentrations in invertebrates and larger consumers. Other elements were present at comparable concentrations in organisms of various trophic levels.

Adams and Johnson (1977) observed an increase in Se levels from water to sediment and from water to zooplankton to fish in western Lake Erie, although the relative contributions of sediment-bound and dissolved Se in bioaccumulation were not investigated. The authors pointed out that the increase in concentration from zooplankton to fish may represent biomagnification or may simply reflect different rates of accumulation by organisms at each trophic level.

An absence of food chain biomagnification has been reported for several metals. Wagemann et al. (1978) observed no biomagnification of As in Northwest Territories lakes. Penrose (1974) reviewed the literature on As in aquatic environments and noted that food chain biomagnification does not occur. The highest Pu concentrations in aquatic organisms generally occur in species feeding or living directly on contaminated sediments and a successive decline in tissue concentrations should be expected with each food chain transfer (Trabalka and Frank 1978). Price and Knight (1978) measured Hg, Cd, Pb and As in sediment, phytoplankton, and clams in two Mississippi lakes and observed generally higher levels of these metals in phytoplankton than in filter-feeding clams, indicating an absence of biomagnification.

Food chain biomagnification of trace metals has also been found lacking in studies on the Great Lakes basin. Copeland and Ayers (1972) determined concentration factors for several trace elements calculated with respect to water and sediment for biota of Lake Michigan and observed that there was no stepwise increase in concentration factors between trophic levels. Hutchinson et al. (1976) studied Ni and Cu in a metal-contaminated watershed draining into Lake Huron near Sudbury. Food chain biomagnification of these elements was not observed.

Factors Affecting Bioaccumulation of Metals from Sediment

Bioaccumulation of metals is influenced by a complex array of biotic and abiotic interactions. The rate and degree of accumulation of metals in organisms exposed to metal-contaminated sediments depend upon such factors as the chemical form and concentration of the metals (Singer 1973, Powers et al. 1977), the particle size and agitation of the sediment and the feeding habits and physiology of the organisms (Delisle et al. 1975). Conceptual problems of toxicant-sediment interactions which must be evaluated in light of chemical and biological availability were discussed by Engler (1979).

Tissue concentrations of metals may vary seasonally. Bryan and Uysal (1978) observed seasonal variations in tissue concentrations of Fe, Zn and Mn in the clam Scrobicularia plana from an English estuary, while tissue Cu levels showed little seasonal variation. In Sandusky Bay, Lake Erie, Hg levels in chironomids were highest in winter and spring and showed no correlation with total Hg in sediments (Skoch and Sikes 1973).

Cases where elevated concentrations of trace metals in sediments do not lead to bioaccumulation have been described. Cross et al. (1970) found no relationship between Mn, Fe and Zn in sediments and tissues of polychaetes in a North Carolina estuary, suggesting that either these organisms can regulate metal uptake or that much of the sediment-bound metal is biologically unavailable. Gillespie (1972) demonstrated that Hg is not bioaccumulated by guppies under certain physicochemical conditions. Neff et al. (1978) observed bioaccumulation of heavy metals from contaminated marine and Lake Erie sediments by four benthic invertebrates. Significant bioaccumulation occurred in only 26.5 percent of their tests. Korda et al. (1977) observed no increase in trace metal levels in sculpins due to the presence of metal-contaminated taconite tailings in areas of Lake Superior.

Several studies have demonstrated the effect of physiological factors on biological uptake of metals from sediment. Delisle et al. (1975) exposed goldfish and catfish to the same contaminated sediment but found different tissue levels of Zn, Pb, Cu and Cd in the two species. Price and Knight (1978) demonstrated that tissue concentrations of heavy metals in the freshwater clams Corbicula manilensis and several unionid species from metal-contaminated sediments in two Mississippi lakes were higher in small clams than in large clams of the same species. Eyres and Pugh-Thomas (1978) defined the "index of accumulation" as mean tissue concentration/mean substrate concentration. The isopod Asellus was shown to have an index of about unity for low substrate Pb levels, but to have a much lower index at higher sediment Pb levels. The authors suggested that Asellus can physically or chemically block Pb uptake or that Pb can be actively excreted. The relationship between sediment Cu levels and tissue concentrations in Asellus and in the leach Erpobdella varied inversely with substrate levels. Zinc was excluded somewhat by Asellus with tissue and substrate levels showing an inverse relationship. Erpobdella actively concentrated Zn at low substrate levels but tissue and substrate levels were approximately equal at high sediment Zn levels. Similarly, in contaminated lakes near Yellowknife, Northwest Territories, concentration factors of As in organisms relative to sediment were found to vary inversely with sediment As concentrations.

Neff et al. (1978) evaluated the bioavailability of heavy metals in polluted sediments from Texas ship channels and from Ashtabula Harbor, Lake Erie to marine and freshwater benthic invertebrates. Variations in bioaccumulation were observed between metals, species, sediments, and salinity. No correlation was observed between specific metal forms and bioaccumulation.

Gillespie (1972) compared the accumulation of various forms of Hg from St. Clair River, Saskatchewan River, and artificial sediments by guppies under aerobic and anaerobic sediment conditions. Under aerobic conditions, metallic Hg was quickly taken up by the fish while Hg in chloride and sulphide forms were only accumulated to a small extent. Under anaerobic conditions, mobilization of Hg from sediment to fish was usually slower.

The availability of sediment-bound metals for bioaccumulation is frequently related to the ability of the metal to desorb from sediment. Cooke et al. (1979) investigated the bioavailability of particulate Cd to the cockle, *Cerastoderma edule*. They observed that Cd bound to biogenic calcium carbonate is readily accumulated while Cd bound to precipitated calcium carbonate is only slightly available and Cd bound to oxides of Fe and Mn is unavailable in short-term exposures. It was concluded that bioavailability is determined by the ability of Cd to desorb from sediment particles. Bioaccumulation of Cd from sediment by the polychaete *Nereis virens* was found to be at the same rate as the dissolution of Cd from sediment into the overlying seawater, suggesting that uptake depended upon the availability of readily dissociated forms (Ray et al. 1980). Similarly, Luoma (1977) concluded that levels of Hg in the deposit-feeding shrimp, *Palaemon debilis* were related to Hg availability from water rather than from sediment.

Luoma and Jenne (1975) studied the bioavailability of sediment-bound Co, Ag and Zn to the deposit-feeding clam, *Macoma balthica*. Bioavailability varied among metals within sedimentary sinks and among sedimentary sinks for a given metal. Bioaccumulation was reduced in several sediment systems which may be common in nature. Little Zn or Co uptake was observed when these metals were co-precipitated with Fe or Mn oxide. Silver, Co and Zn were all bioaccumulated from detrital organics and Ag was taken up from Fe oxide precipitate. Cobalt and Zn associated with biogenic carbonates were more easily accumulated than the same metals associated with other sedimentary components. Similarly, Ag uptake from biogenic and synthetic carbonates exceeded uptake from Fe oxides and detrital organics. Sedimentary pools from which bioaccumulation occurred most readily showed the greatest rates of metal desorption.

Several other investigations have indicated that the total or bulk sediment concentration of metals is generally unavailable for tissue accumulation (Bertine and Goldberg 1972; Skoch and Sikes 1973; Brannon et al. 1976; Luoma 1977; Turekian 1977).

Effects on Density and Diversity of Organisms

Trace metal contaminants have been linked to changes in structure of aquatic communities. Most studies have shown that metal-contaminated systems support simplified communities due to the elimination of more sensitive species.

Metal-contaminated coal ash reduced the diversity of the biological community in a South Carolina watershed (Guthrie et al. 1974, Cherry and Guthrie 1975, Cherry et al. 1979). Primary and secondary consumers were particularly affected. A limited number of invertebrates, primarily chironomids and odonates, and only one fish species (mosquitofish, Gambusia affinis) were present. Aquatic bacterial populations were apparently unaffected (Guthrie et al. 1974). After completion of a coal ash retaining system to permit settling of metal-contaminated solids, diversity and abundance of aquatic organisms in the receiving system recovered (Cherry et al. 1979).

In the River Irwell (England), which has high levels of Pb, Zn and Cu in the sediment, Eyres and Pugh-Thomas (1978) concluded that the absence of fish and the paucity of invertebrates can be at least partially attributed to heavy metal contamination, particularly since high dissolved oxygen levels are maintained.

In the lower Hudson River (New York), Occhiogrosso et al. (1979) sampled benthic invertebrates and sediments in the vicinity of a source of Cd contamination. The authors demonstrated that benthic densities were lowest where levels of Cd in the sediment were highest. Since other parameters (organic carbon, particle size distribution, and total exposure) were very similar at all stations, the decrease in benthic density was attributed to heavy metal contamination.

Wagemann et al. (1978) examined the benthic communities of As-contaminated lakes in the Northwest Territories and found that clams, mayflies, amphipods, and leeches were conspicuously absent in lakes with the highest As concentrations in sediment.

In an Indiana lake with sediments contaminated by Cd, Zn and Cr, Wentsel et al. (1977) found that larvae of the midge *Chironomus tentans* were absent in the most contaminated areas, but increased in abundance in areas having lower sediment metal concentrations. Conversely, densities of the oligochaete *Limnodrilus* sp. were highest in the most heavily impacted areas of the lake. The authors suggested that elimination of competitors and predation in contaminated areas probably accounted for high oligochaete densities.

Moore (1979) studied benthic densities and diversities in a contaminated bay of Great Slave Lake, Northwest Territories, where sediments contained elevated concentrations of As, Hg, Pb, Cu and Zn from mining waste. Benthic collections were made in areas of heavy contamination, moderate contamination, and negligible contamination. Species diversity was an ineffective indicator of contamination, but a strong negative correlation between the concentrations of sediment-bound metals and total abundance of benthic macroinvertebrates was shown. Winner et al. (1975) studied benthic diversities in a metal-contaminated temperate stream and also found that diversity indices were inadequate for delineating the effects of heavy metals on the benthic community.

Moore et al. (1979) attempted to separate the effects of sediment and water contamination by heavy metals on benthic invertebrate communities in two Northwest Territories lakes. Since the concentrations of heavy metals in two adjoining lakes were statistically alike, the authors suggested that the lower density and diversity in one lake were attributable to higher levels of dissolved contaminants (primarily Cu and cyanide) observed in this lake.

A study by Sweeney et al. (1975) on benthic invertebrates at a Lake Erie disposal site for dredged materials from Cleveland Harbor demonstrated that sediments contaminated by heavy metals as well as other pollutants supported a low species diversity and a high ratio of oligochaetes to total organisms relative to the surrounding sediments.

In an area of Lake Superior contaminated by Cu-contaminated mine tailings, densities of the amphipod Pontoporeia hoyi showed a significant negative correlation with sediment Cu concentrations (Kraft 1979).

In contrast to most studies, Hutchinson et al. (1976) found no major differences "in populations or seasonality of the biota" between metal-impacted and uncontaminated streams of a watershed draining into Lake Huron near Sudbury.

Biological Indicators of Metal Contamination

Few studies have described biological indicators of metal pollution in freshwater environments. Surber (1959) presented data on invertebrates above and below an electroplating plant discharge into a Michigan stream. Chironomids increased from 7.5 percent of the total individuals collected upstream of the discharge to 96 percent downstream from the discharge. The work of Winner et al. (1980) on metal-contaminated streams in Ohio suggested that the relative numbers of chironomids, caddisflies and mayflies may indicate various degrees of heavy metal stress. The authors concluded that only chironomids survive in the most polluted area, chironomids and caddisflies are both abundant in zones of moderate metal pollution, and caddisflies and mayflies predominate in stream sections subject to little metal pollution.

Lang and Lang-Dobler (1979) statistically related oligochaete assemblages and sediment chemical parameters in Lake Geneva (Switzerland) to assign biological index values describing various types and levels of sediment contamination. The assemblages characteristic of various metallic contaminants were *Peloscolex ferox*, *Potamothrix hammoniensis* and *Limnodrilus claparedeanus* for Cd; *Psammoryctides barbatus* for Zn: *Potamothrix vejdovskji* for Hg; and *Peloscolex velutinus* and *Stylodrilus lemani* for Mn.

IMPACT OF PERSISTENT ORGANIC-CONTAMINATED SEDIMENT ON BIOTA

Biological Uptake

While persistent organic contaminants may pose immediate threats to organisms through toxicity, perhaps the most important impact of such substances is biological uptake resulting in potential toxicity to man through the consumption of contaminated aquatic organisms.

Many chlorinated hydrocarbons are both toxic and persistent and some tend to be readily accumulated by organisms in which they may exert long-term chronic effects. A summary of the carcinogenic and mutagenic properties of organohalogens provided by Young et al. (1979) indicated that these substances represent a particularly dangerous family of chemicals.

In this review, "bioaccumulation", "biomagnification" and "bioconcentration" are used as outlined in the corresponding section on trace metals (Impact of Metal-Contaminated Sediment on Biota).

Some favour food chain uptake as the predominant route for the uptake of persistent organics to upper trophic level biota (Sodegren 1973; Norstrom et al. 1976) while others feel that direct uptake or partitioning via respiratory activity is most important (Branson et al. 1975; Sanborn et al. 1976; Chiou et al. 1977). Few have examined the role of sedimentary sources in biological uptake.

The bioavailability of sediment-bound persistent organics depends on properties of both contaminants and sediments (Weber 1977). Compounds that are weakly adsorbed to the surfaces of sediment particles are readily desorbed and dissipate in the aqueous environment. Pesticides such as diquat and paraquat that are adsorbed onto the internal surfaces of expanding clays are not desorbed and are unavailable to biota. Contaminants having low solubilities normally associate with the surface of lipophilic sediment and are in equilibrium with the surrounding aqueous phase. Desorption tends to increase bioavailability of these substances.

Bioaccumulation

Microbes

Due to their ability to bioaccumulate toxic organic substances, bacteria influence the transfer of organics to the sediments. Leshniowsky et al. (1970) and Pfister et al. (1971) demonstrated that floc-forming bacteria isolated from Lake Erie concentrate organochlorine aldrin occurring in association with suspended colloids in the Lake Erie water column. The former authors stated that the bioconcentration factor in these bacteria over background levels in the water column was 625 times.

Bacteria also metabolize toxic organic substances to daughter products which may differ in toxicity and bioaccumulation potential from their parent compounds (Guenzi & Beard 1967; Lotse et al. 1968; Miles 1977).

Primary Producers

Phytoplankton bioaccumulate toxic organics through the process of adsorption (Hiraizumi et al. 1979). However, research on bioaccumulation of sediment-bound organic contaminants by phytoplankton has apparently not been carried out.

Rooted aquatic macrophytes may accumulate organic contaminants from sediment. Gallagher et al (1979) demonstrated that the salt marsh plant Spartina alterniflora bioaccumulates the hydrophobic pesticide toxaphone from sediment through the roots.

Benthic Invertebrates

Apart from studies including benthos as one ecosystem component in the examination of contaminants in sediment, water and various biotic components of freshwater ecosystems (e.g., Haile et al. 1975; Sarkka et al. 1978), little is known about bioaccumulation of toxic organic substances by freshwater benthos.

In contrast, studies of bioaccumulation by marine invertebrates are numerous and include both laboratory and field observations. Most workers dealing with bioaccumulation by marine invertebrates from sediment have examined the organochlorine PCB. Shuba et al. (1978) exposed estuarine clams (Rangia) and shrimp (Palaemonetes) to a suspended sediment contaminated with PCB. Test animals accumulated an average of five to ten times as much PCB residue as control animals after 14 days. Langston (1978) found that PCB's associated with particles of aluminum oxide were accumulated by Macoma balthica. Elder et al. (1979) demonstrated the accumulation of PCB from contaminated sediments by the polychaete, Nereis diversicolor and suggested that uptake was due either to sediment ingestion or to cuticular absorption from interstitial water. Polikarpov et al. (1979) demonstrated that N. diversicolor can accumulate PCB both from sediment and from seawater. Although the bioconcentration factor from water was substantially higher than from sediment (800 vs. 3.5), it was concluded that sediments are the major source of PCB for these worms under existing environmental concentrations in the Mediterranean Sea. Courtney and Langston (1978) exposed the polychaetes, N. diversicolor and Arenicola marina to concentrations of PCB that might be expected in contaminated sediment and seawater. Similar levels of residues were accumulated in either case. Courtney and Langston concluded that in less polluted conditions where PCB is undetectable in solution, sediments may be the primary source of bioaccumulated PCB. Nimmo et al. (1975) concluded that pink shrimp in Escambia Bay obtain most of their PCB body burden by uptake from seawater. PCB levels in sediments and bivalves in Raritan Bay at the mouth of the Hudson River estuary were reported by Stainken and Rollwagen (1979). Although the mode of bioaccumulation was not determined, the highest tissue levels generally occurred at stations where sediment contaminant levels were highest. The uptake of 11 sediment-bound chlorinated hydrocarbons from suspended sediment by five species of freshwater and marine fish and invertebrates was studied by Peddicord and McFarland (1978). Limited uptake occurred in two cases and possibly in a third. PCB and DDE were bioaccumulated by the marine mussel Mytilus edulis, while DDE was possibly accumulated to a small degree by Mytilus californianus.

Fish

Few studies have demonstrated the relative importance of sediment and water sources to bioaccumulation of toxic organic compounds in fish. Halter and Johnson (1977) studied the uptake of PCB's by fathead minnows, Pimephales promelas from contaminated sediment. Fish screened from direct access to the sediment accumulated PCB's six times more slowly than those permitted to forage in the polluted substrate. The activity observed in the unscreened fish of passing contaminated sediment into the bucial cavity and then expelling it was thought to cause elevated dissolved PCB levels in the buccal cavity leading to branchial uptake.

The uptake of mirex by the hogchoker, Trinectes maculatus in a contaminated sediment-water system was investigated by Kobylinski and Livingston (1975). The authors observed that considerably more mirex was gained by the fish than was lost from the water and concluded, therefore, that a significant amount of mirex was absorbed from sediment.

Kent and Johnson (1979) suggested that sediments may be an important source of organochlorines to Utah suckers in American Falls Reservoir, Idaho. This species accumulated higher levels of organochlorines than other fish and was the only species containing detectable PCB residues in the reservoir. These high organochlorine levels in suckers were attributed to exposure to sediment-bound contaminants during feeding activities.

Dexter et al. (1978) found that PCB residues in sediment were generally proportional to tissue levels found in carp over a section of the Mississippi River in Minnesota.

Tissue concentrations of organochlorine residues in Great Lakes fish have been reported by several workers. Although most of these investigations do not differentiate between the relative importance of dissolved and sediment-bound forms in bioaccumulation, high levels in fish have been reported in areas having relatively high sediment contaminant loads. Summaries of some of these investigations follow.

Delfino (1979) discussed the occurrence of PCB's in Lake Michigan fish and also discussed the bioenergetic model developed by Weininger in his Ph. D. thesis accounting for the accumulation of PCB by Lake Michigan lake trout. This model predicted that greater than 50 percent of the PCB's in adult lake trout had previously cycled through sediments in the lake.

Higher concentrations of organochlorine pesticides in fish were found by Frank et al. (1974) in areas of higher sediment contaminant levels in the Muskoka Lakes which drain into Georgian Bay. Concentrations in fish tissue also varied with feeding habits and increased with increasing fat content and age.

Levels of DDE, HEOD and PCB in Lake Superior sediments measured by Frank et al. (1980) were considerably lower than levels observed in downstream lakes (Frank et al 1977, 1979a, b) which conforms with lower residues found in Lake Superior fish than found in other Great Lakes (Reinert 1970; Reinke et al. 1972; International Joint Commission 1976; Frank et al. 1978).

Reinert (1970) reported concentrations of organochlorine pesticides in fish of all of the Great Lakes and found that residues in Lake Michigan fish were two to seven times greater than observed in fish of the other Great Lakes. More details on organic contaminants in fish, sediments, and waters of Lake Michigan were provided by Schacht (1974). Reinke et al. (1972) reported organochlorine residues in commercially caught Great Lakes fish. Carr et al. (1972) reported organochlorine residues in six fish species from Lake Erie. Organochlorine concentrations in fish, water, and sediment of Great Lakes watersheds were provided by Miles and Harris (1971, 1973, 1978) and Frank et al. (1974).

While reports of organochlorine levels in marine fish are numerous, few of these studies have discussed the role of sediment-bound contaminants in bioaccumulation. Young et al. (1977) and McDermott et al. (1976) measured levels of PCB's and DDT in Dover sole, Microstomus pacificus, following reductions in wastewater inputs into southern California waters. Despite significant decreases in inputs from municipal wastes, levels in Dover sole tissue did not decline, apparently due to sediment reservoirs of these hydrocarbons.

Biomagnification

Persistent organic contaminants may show step-wise increases in tissue concentration with increasing trophic level due to food chain biomagnification.

Haile et al. (1975) and Sarkka et al. (1978) examined the concentrations of chlorinated hydrocarbons in ecosystem components of Lake Ontario and a Finnish lake, respectively. In both cases, step-wise increases with trophic levels were observed. Much of the apparent biomagnification effect observed with chlorinated organics might still occur without food chain transfer since these compounds tend to accumulate in lipids and many top predators have relatively large fat deposits (Hamelink et al. 1971; Portmann 1975). Portmann (1975) determined that if DDT concentration were calculated on a lipid rather than on a wet weight basis, biomagnification along the phytoplankton - seal food chain would be reduced from several orders of magnitude to less than one order. The data of Elder and Fowler (1976) and Cahn et al. (1977) suggest that PCB's may not be biomagnified from phytoplankton to fish. Pearson and McConnell (1975) found no clean evidence for the biomagnification of C_2-C_2 aliphatic organochlorines such as perchloroethylene.

Factors Affecting Bioaccumulation of Persistent Organics from Sediment

Investigations of factors influencing the biological uptake of toxic organic contaminants are few. In general, dissolved forms of contaminants are more biologically available than particulate forms (Pionke 1977). For instance, the bioconcentration factors for PCB from seawater and sediment were 800 and 3.5 respectively, for the polychaete Nereis diversicolor (Polikarpov et al. 1979). Therefore, physicochemical properties of contaminants, sediments, and water which govern sediment-contaminant interaction (see Section 2) influence bioavailability.

Levels to which hydrophobic, lipid-soluble compounds are accumulated depend on such factors as the lipid-water partition coefficient of the contaminant and on the amount of lipid in the organism. To illustrate this point, Ernst (1977) equilibrated Mytilus edulis with equal concentrations of chlorinated hydrocarbons and found that concentration factors in the mussels were inversely proportional to the water solubility of the compounds.

Olsson et al. (1978) demonstrated that organochlorine levels may vary seasonably in fish. PCB levels in roach (Leuciscus rutilus) from a Swedish lake were determined up to 12 times annually for seven years. Levels peaked in spring and then decreased rapidly back to a base level. This PCB pulse was attributed to the flushing of contaminated sediments into the lake during spring run-off, although it was also suggested that reproductive activities may also play a role in increase uptake in spring.

Kaiser (1979) deduced that water quality conditions, particularly turbidity and phytoplankton density, inversely affect the amount of organic contaminants available for biological uptake in the Great Lakes. Concentrations of PCB in fish, herring gull eggs and sediments were compared in Lake Superior, Lake Erie and Lake Ontario. It was shown that the concentrations and total amounts of contaminant in fish and herring gull eggs were similar for each species in all three lakes while the estimated total PCB loads in sediments differed considerably, with the highest concentrations occurring in Lake Erie. Kaiser postulated that the high phytoplankton density in Lake Erie results in the quick adsorption of PCB entering the lake and in subsequent sedimentation. In less productive lakes such as Lake Superior and Lake Ontario, higher proportions of the phytoplankton biomass are ingested by primary consumers, apparently leading to a greater proportion of the total PCB load entering the food chain.

Effects on Biological Communities

Although organohalogens represent a particularly dangerous family of carcinogenic and mutagenic chemicals which occur widely in aquatic ecosystems (Young et al. 1979), apparently no studies have focused attention on community responses to chronic contamination by persistent organics. This may be attributable largely to the fact that industrial, municipal, and agricultural sources usually introduce a myriad of organic and inorganic pollutants which tend to obscure possible impacts of persistent organics.

Spills of toxic organic chemicals can have catastrophic effects on aquatic communities. For instance, the release of pentachlorophenol into a watershed in Mississippi resulted in the extensive to total destruction of the fish community in a downstream reservoir (Pierce et al. 1977). Flannagan et al. (1979) induced a massive drift of benthic organisms over a 400 km section of the Athabasca River (Alberta) by injecting methoxychlor into the river at a concentration of 0.3 mg/l for 15 min. While such catastrophic impacts may occur when substantial amounts of dissolved contaminants are introduced immediately into an aquatic environment, the impacts of long-term exposure to sediment-bound organic contaminants on community structure would normally be less drastic.

Results of toxicity tests indicate that chronic exposure to persistent organics is detrimental to individual species. Hansen et al. (1971) found that exposure to PCB increased the susceptibility of pinfish, Lagodon rhomboides, and spot, Leiostomus xanthurus, to disease. Rubinstein (1979) demonstrated that the polychaete, Arenicola cristata, reduces its substrate re-working activity when exposed to low levels of kepone. Birge et al. (1977) found that the frequency of teratogenesis increases appreciably in surviving fish hatched over contaminated sediments. Such sublethal effects would be expressed in nature by changes in community structure.

METHODS FOR MONITORING BIOLOGICAL UPTAKE OF SEDIMENT BOUND CONTAMINANTS

BIOACCUMULATION-LABORATORY METHODOLOGY

The bioavailability of sediment-bound contaminants varies not only with the myriad of physicochemical properties governing contaminant concentrations and distributions in sediment, but also with specific physiological characteristics of the organisms concerned. Several studies have been undertaken to determine the bioaccumulation potential of various contaminants in both freshwater and marine sediments. A review of field methods, laboratory tests, and toxicity data concerning toxicant bioaccumulation from sediment was provided by Engler (1979).

Primary Producers

Laube et al. (1979) studied the mobilization and accumulation of sediment-bound Cd and Cu by freshwater algae (Anabaena sp. and Ankistrodesmus braunii) in the laboratory. Algae were placed in dialysis sacs and suspended in a system of circulating water and contaminated sediment. The distribution of metals in water, sediment and algae was measured at selected intervals for up to 72 hours. The authors demonstrated indirect biological uptake from the sediments and concluded that algae may play an important role in the biomobilization of sediment-bound heavy metals in natural systems.

Bioaccumulation of Pb from sediment by the aquatic macrophyte, Vallisneria americana, and by phytoplankton was examined by Milne and Dickman (1977). The authors grew these organisms in aquaria containing artificially-contaminated and natural sediments and observed that biologically available Pb from sediment was concentrated by the test organisms.

Uptake of the radionuclides Ru-106/Rh-106 and Cs-137 by the marine alga Cryptonema sp. was measured by Ueda et al. (1978). The alga was exposed to contaminated, filtered seawater and to a contaminated sediment-seawater system and tissue content was measured periodically for up to two weeks. The experimental design permitted the demonstration of the relative importance of sediment and seawater on biological uptake.

In a study sponsored by the Dredged Material Research Program of the U.S. Army Engineer Waterways Experiment Station, Patrick (1977) investigated the uptake of various heavy metals from sediments by the marsh plants Spartina alterniflora, Spartina cynosuroides and Distichlis spicata. The authors maintained the roots of these plants in soil suspensions in sealed containers where pH, redox, and salinity could be controlled in order to determine the influence of these parameters on biological uptake. The experimental techniques which were successful, as well as many approaches which were not successful, were outlined and advantages and limitations of these techniques were discussed.

Benthic Invertebrates

Few laboratory investigations of bioaccumulation from sediments by freshwater benthic invertebrates have been documented. Derr and Zabik (1972) applied DDE in solution to a flow-through water-sediment system and measured uptake by various life history stages of the midge Chironomus tentans. Trabalka and Frank (1978) stocked Pu-contaminated sediment-water microsms with adult Chironomus reparius which mated and produced a larval generation. Plutonium concentrations in midge larvae, sediments, and water were measured periodically to determine patterns of Ph distribution. Neither experimental designs permitted differentiation between uptake from substrate and from water.

Neff et al. (1978) conducted laboratory experiments on metal uptake by the freshwater invertebrates Tubifex sp. and Palaemonetes kadiakensis and by the marine invertebrates Rangia cuneata and Neanthes arnaceodentata using contaminated marine sediments from Texas waters and contaminated freshwater sediments from Ashtabula Harbor (Lake Erie). The experiments examined variations in bioaccumulation of ten metals among species, metals, sediments, and salinities.

Laboratory studies on bioaccumulation of contaminants from sediment by benthic marine organisms are numerous. Many of the basic principles and procedures described may be applied to freshwater studies.

Polikarpov et al. (1979) and Elder et al. (1979) exposed Nereis diversicolor to PCB-contaminated sediment and measured uptake and subsequent depuration until equilibrium levels were reached (several weeks). Their methods did not permit the determination of the mode of uptake.

Bryan and Hummerstone (1971) monitored metal uptake by the polychaete Nereis diversicolor from contaminated estuarine sediments. Levels of Cu in exposed worms were correlated with sediment Cu concentrations. Concentrations of Zn, Pb, Mn and Fe were relatively constant regardless of sediment concentrations, suggesting the Nereis is capable of regulating uptake of these metals.

Investigations on PCB uptake by Nimmo et al. (1971) and Courtney and Langston (1978) accounted for the bioavailability of dissolved and sediment-bound PCB's. The former authors exposed the shrimp, Pennaeus duorarum and the crab, Uca mimax to contaminated sediment and seawater. The latter investigators mixed PCB-seawater solutions with various sediment particle sites to produce different sediment-contaminant combinations. Biological uptake in contaminated systems and subsequent depuration in uncontaminated systems by the polychaetes, Nereis virens and Arenicola marina were monitored over several days.

Bioaccumulation of Zn, Co and Ag by the deposit-feeding clam, Macoma balthica from various artificial substrates were reported by Luoma and Jenne (1976, 1977). They corrected for the absorption of metals leached from the sediments by enclosing control animals in dialysis sacs above the sediments. Cooke et al. (1979) investigated the bioavailability of Cd bound to various forms of calcium carbonate to the cockle, Cerastoderma edule. In both studies, the authors demonstrated that bioavailability of metals depended on sediment type.

Ueda et al. (1978) and Ray et al. (1980) also investigated the uptake of trace elements in simple seawater-sediment-organism systems. The former authors studied uptake of the radionuclides Ru-106/Rh-106 and Cs-137 by the bivalve Gomphina melanaegis in contaminated sediment-seawater systems. Ray et al. (1980) measured tissue Cd levels in the polychaete, Nereis virens maintained in contaminated sediment-seawate systems over 25 days and then measured subsequent depuration rates over 75 days. In both investigations, bioaccumulation from sediment and from seawater were assessed by maintaining control animals in contaminated seawater alone.

The feeding strategy of experimental animals is an important factor in biological uptake. Phelps and Myers (1977) investigated heavy metals in animals which feed from the overlying water column, at the sediment water interface, and below the sediment-water interface in contaminated and uncontaminated areas of Narragansett Bay, Rhode Island. The burrowing subsurface feeders showed the poorest correlation between sediment and body burdens of heavy metals. Water column feeders and interface feeders contained the highest incidence of heavy metal contamination, suggesting that these groups are best for biological monitoring of heavy metals in sediment. Anderson et al. (1978) studied the biological uptake of petroleum hydrocarbons from sediments in an experimental system using a filter-feeding clam (Protothaca staminea), a detritus-feeding clam (Macoma inquinata), and a sediment-ingesting sipunculid (Phascolosoma agassizii). The authors found that the two deposit-feeding species accumulated considerably higher levels of petroleum hydrocarbons than did the filter-feeding species.

Many other studies have been conducted on the biological uptake of contaminants by benthic invertebrates from petroleum-contaminated sediments. Descriptions of some of these investigations follow.

Rossi (1977) exposed the polychaete, Neanthes arenaceodentata to seawater and sediment contaminated with water-soluble petroleum hydrocarbons and radio-labelled naphthalenes and observed uptake over four weeks. The experiment suggested that hydrocarbons bound to sediment and to particulate organic matter are less available to the worms than those in solution.

The investigations of Anderson et al. (1977) and Roesijadi et al. (1978) demonstrated the bioaccumulation of radio-labelled naphthalenes and by the deposit feeders *Phascolosoma agassizii* and *Macoma inquinata*, respectively, and showed that uptake due to ingestion of sediment was insignificant while dissolved fractions were biologically available.

Anderson et al. (1979) exposed the amphipod *Anonyx laticoxae* to whole oil in sediment and to water extracts of oil under both static and flowing conditions for up to 27 days. Sediment-bound petroleum hydrocarbons displayed relatively low bioavailability and uptake appeared to be primarily from interstitial water and from water column contamination. These conclusions were based on the types of hydrocarbons found in tissue, water, and sediment.

A series of laboratory investigations on bioaccumulation of contaminants from petroleum-contaminated sediments was reported by Anderson et al. (1978). The uptake of various sediment-bound and dissolved hydrocarbons by *Macoma inquinata* was observed over seven days. One group of clams was placed on the test sediments while another group was suspended above the sediment in a mesh basket. After exposure, half of the clams from each group were analyzed for hydrocarbons while the remaining clams were placed in clean seawater for 24 hours to permit purging of the gut before analysis. Concentrations of contaminants in sediments, water, and tissues were measured and magnification factors were calculated. This technique distinguished uptake from water and sediment. *Macoma inquinata* was also used to study uptake of radio-labelled petroleum hydrocarbons from sediment in simulated intertidal conditions. Suspended contaminated or control sediments were added to trays of sand holding test animals at "high tide". Seawater was completely drained from the trays through mesh bottoms at "low tide". At each sampling interval for up to six weeks, a group of clams and a sediment sample were removed for analysis of hydrocarbon content. Contaminant levels in food were accounted for by placing subsamples of clams in clean seawater for 24 hours to allow the purging of gut contents. The authors carried out an experiment on indirect and direct bioaccumulation of petroleum hydrocarbons by the polychaete *Abarenicola pacifica* from sediments contaminated only in a shallow surface layer (over a layer of clean sediment) and from sediments contaminated homogeneously with depth. This worm lives and feeds in an L-shaped burrow and the horizontal portion of the burrow houses the anterior portion of the worm. The experiments were designed such that the horizontal portion of the burrow was either in a contaminated or an uncontaminated layer. Naphthalene levels were measured periodically in sediment, worm feces, and in various worm tissues. On the basis of their data, the authors conjectured that A. pacifica can accumulate petroleum hydrocarbons by direct contact with water and surrounding sediment and through ingested material. The influence of the presence of petroleum contamination in sediment on the uptake of heavy metals from detritus was also investigated. The test animals Phascolasoma agassizii and Macoma inquinata were divided into two groups. One group was exposed to metal-contaminated oiled sediment or to metal-contaminated unoiled sediment while a second group was maintained over the contaminated sediments in mesh baskets. Direct access to the sediment resulted in increased uptake of metals while the presence of oil had no effect on bioaccumulation.

Dredging activities result in the suspension of sediments at both the dredging and disposal sites. Peddicord and McFarland (1978) conducted laboratory experiments to investigate bioaccumulation of toxic substances from suspended sediment. Marine invertebrates were exposed to contaminated sediments at various salinities. Tissue levels of nine metals and the organohalogens chlordane, heptachlor, dieldrin, aldrin and endrin, DDT and its derivatives and PCB's were monitored over 21 day exposures. Suspended uncontaminated sediments were used in controls. Of 100 species-salinity-contaminant combinations, significant uptake occurred in less than one fourth of the cases.

Emerson (1976) mixed combinations of contaminated Los Angeles Harbor sediments with seawater and observed biological uptake by the polychaete, Capitella capitata from the supernatant elutriates over 28 days. Uptake of various contaminants did not correlate with sediment contamination level, apparently due to the binding of elutriated contaminants with organic materials in the suspended solids fraction. The silt content of the most highly contaminated sediments was generally higher than in other sediments tested, leading the author to postulate that higher suspended silt levels in the elutriate would result in adsorption of contaminants and decreased bioavailability.

Fish

As in general bioassay applications, fish retain their importance in laboratory studies on bioaccumulation of contaminants, including those associated with sediments.

Delisle et al. (1975) investigated the rate and degree of uptake of Cu, Pb, Zn and Cd from grossly contaminated sediment by goldfish (Carassius auratus) and catfish (Corydoras aeneus). Concentrations in dry, whole fish tissue from test and control aquaria were analyzed initially and at one-month intervals for four months. Although the method did not distinguish between uptake from sediment-bound and desorbed sediment, the technique provided a useful indication of bioaccumulation potential of the sediments.

Since physicochemical factors can exert a great influence on the form and bioavailability of trace metals in sediments, Gillespie (1972) measured the uptake of various forms of Hg (elemental, mercuric chloride and sulphide, methylmercury) from both aerobic and anaerobic sediment by guppies (Poecilia reticulata). Mercury was dispersed into test sediments by sonic oscillation. Anaerobic sediments were prepared by keeping contaminated sediments in tightly capped jars until anoxic conditions were established. Jars were placed in test aquaria and their caps removed. A mesh-covered top on the jars inhibited oxygen influx. Uptake by guppies was observed over periods of up to 150 days.

Halter and Johnson (1977) studied PCB desorption from contaminated sediment and subsequent uptake by fathead minnows (Pimephales promelas). Acetone solutions of PCB were applied to sediment in various concentrations. Fish were exposed to various PCB levels in sediments and were either allowed access to the substrate or were separated from the bottom by screens. Tissue PCB loads were measured over 32 days. The rate of dissolution of PCB's from sediment was also measured. Although fish having direct access to the sediments accumulated PCB residues at six times the rate of those denied access, chromatograms of Aroclor 1254 residues in water, sediment, and in the two groups of fish indicated that PCB was taken up primarily from water in all cases. The authors attributed higher uptake in fish having access to the bottom to grazing at the sediment surface resulting in elevated levels of dissolved PCB's in the buccal cavity and subsequent uptake via the gills.

Kudo and Mortimer (1979) applied a similar approach to monitor uptake of Hg from sediment and from water containing Hg desorbed from sediment by freshwater fish. They also found a much greater rate of bioaccumulation when fish had direct access to the substrate.

The bioaccumulation of mirex from sediment by the hogchoker, *Trinectes maculatus* was studied by Kobylinski and Livingston (1975). This species was selected as an experimental animal since it is carnivorous and does not likely ingest bottom material directly. Thus, accumulation of the contaminant could be observed without the complication of direct consumption. Sand was artificially contaminated with three concentrations of mirex and then covered with freshwater. Fish were exposed to contaminated static and flow-through systems for a month and mirex levels in sediment, water and fish tissue were analyzed weekly. The authors observed that considerably more mirex was gained by the fish than was lost from water and, therefore, concluded that the fish probably absorbed a significant amount of mirex from the sediments.

Fujiki et al. (1977) investigated methylmercury uptake by the red sea bream, *Chrysophyrs major* from seawater, food, and from seawater-sediment systems to determine the relative importance of various methylmercury sources to biological uptake. Experimental results indicated that dissoved methylmercury was the primary source for bioaccumulation.

McCain et al. (1978) exposed the English sole, *Paraphyrs vetulus* to experimentally-oiled sediments over four months in the laboratory and showed that alkanes and aromatic hydrocarbons were readily accumulated. Petroleum hydrocarbon distribution among tissues and pathological consequences of uptake were also examined.

The injestion, absorption, and retention of radio-labelled Mn, Fe, Co and Zn from sediments by the marine demersal fish Kareius bicoloratus were studied by Koyanagi et al. (1979). Contaminated sediment was administered orally in gelatine capsules to anaesthetized fish and radioactivity in whole fish was measured periodically over one week to develop a retention curve. The experiment demonstrated that the incidental ingestion of sediment during feeding may be an important mode of bioaccumulation in bottom-feeding fishes.

Uptake of contaminants by fish from elutriates of contaminated Los Angeles Harbor sediments was studied by Chamberlain (1976) and Brewer (1976). Chamberlain (1976) prepared elutriates of contaminated sediments (as outlined by USEPA/U.S. Corps of Engineers Technical Committee on Criteria for Dredged and Fill Material, 1977) and exposed white croaker, Genyonemus lineatus, to the elutriates over 14 days in order to measure acute toxicity and biological uptake. Brewer (1976) prepared elutriates by combining seawater and sediment in 4:1 to 100:1 ratios, mixing them thoroughly for 30 minutes and allowing them to settle for 60 minutes. Bioassays were conducted with northern anchovy eggs, larvae, and adults over 96 hour periods in aerated elutriates. In another experiment, seawater and contaminated sediment were mixed in aquaria containing anchovies. In all cases, tissue levels in surviving animals were measured.

IN SITU MONITORING OF BIOACCUMULATION

The monitoring of bioaccumulation potential of contaminated sediments in situ has been undertaken by several workers. However, such studies are frequently designed so that test organisms accumulate contaminants from all environmental sources, including dissolved pollutants and contaminated food. The USEPA/U.S. Corps of Engineers Technical Committee on Criteria for Dredged and Fill Material (1977) recommended that in situ monitoring of bioaccumulation by caged animals be carried out in conjunction with laboratory studies.

Mayes et al. (1977) grew the aquatic macrophyte, Elodea canadensis in wooden containers placed in a metal-contaminated lake and in an uncontaminated lake. Plants were rooted in sediments from each lake in each of the two environments. Since the plants were exposed to all combinations of contaminated and uncontaminated sediment and water, the experimental design permitted differentiation between uptake from sediment and from water and, therefore, the relative importance of shoots and roots in bioaccumulation were demonstrated.

Rudd et al. (1980) investigated bioaccumulation by several aquatic organisms in experimental enclosures in Hg-contaminated Clay Lake, Ontario. The influence of Se and herbicide concentration on the compartmentalization of Hg and Se among the biotic and abiotic components of the system were monitored.

Freshwater unionid mussels are becoming increasingly popular as monitoring tools for the assessment of environmental contamination. Merlini et al. (1978) stated that the Unionidae are excellent indicators of metal pollution because they have an affinity for heavy metals and because they are sedentary and reflect the immediate environment of water and sediment. The authors used unionids to evaluate river pollution from an electroplating industry before and after installation of a wastewater purification system.

Results of a study by Flannagan et al. (1979) suggested that caged animals may be inadequate indicators of environmental damage. They released methoxychlor into the Athabasca River (Alberta) and found that caged animals accumulated significantly different residues from those in natural populations.

BIOMAGNIFICATION

Laboratory studies on biomagnification of contaminants derived from sediments are few. Knowledge of the potential for biomagnification of sediment-derived pollutants should be included in site-specific evaluations of environmental impacts of contaminated sediment if significant bioaccumulation occurs. Since bioaccumulation potential is often assessed using primary consumers and detritivores as test organisms, information is also required on transfer to higher trophic levels which may serve as food sources for man. Terhaar et al. (1977) devised an experimental system capable of distinguishing between bioaccumulation and biomagnifications using phytoplankton, Daphnia magna, freshwater mussels, and fathead minnows (Pimephales promelas). The system described permits repeated simultaneous sampling of water and the various trophic levels over an extended period. Modification of this method to incorporate test sediment could be useful in the development of biomonitoring techniques for the assessment of sediment pollution.

RECOMMENDED U.S. ENVIRONMENTAL PROTECTION AGENCY PROCEDURES

The U. S. Environmental Protection Agency/U.S. Corps of Engineers Technical Committee on Criteria for Dredged and Fill Material (1977) published an implementation manual for the ecological evaluation of dredged material disposal. Ocean disposal criteria outlined under Section 103 of Public Law 92-532 require that the potential for bioaccumulation of contaminants from dredged material be evaluated in the technical assessment of permit applications (Peddicord 1977).

The field assessment procedure outlined is valid only where there is an historical precedent for the proposed operation being evaluated (Peddicord 1977). Therefore, the technique is applicable in the case of maintenance dredging where the quality of the sediment to be dredged is considered unchanged since the last dredging and disposal operation. Also, the proposed site must be the same site at which the dredged material concerned has been previously disposed or must have a similar sediment type and biological community as previous disposal sites. Under these conditions, it is possible to assess bioaccumulation by animals that have spent major portions of their lives in or on a sediment very similar to the sediment proposed for dredging under the physical and chemical conditions actually occurring at the disposal site.

The test species must occur in sufficient numbers for sample analysis at disposal sites. These species should be relatively immobile to ensure that body burdens of contaminants at the time of collection are related to the collection location. Tissue levels in organisms from at least three stations within the disposal site boundaries and at least six stations outside of the disposal site must be determined. Sediment characteristics at all sites must be similar. Three of these stations outside of the disposal area should be in the path of sediment transport from the disposal site to provide an indication of contaminant transport. The other three stations should be in a direction opposite that of the net bottom transport to serve as control sites. _In situ_ bioaccumulation in caged organisms may be investigated in combination with laboratory studies.

Animals to be analyzed for tissue contamination are transported to the laboratory and placed in clean water of "nonstressful" temperature and dissolved oxygen conditions. The animals are provided with no food or sediment. Fecal matter is siphoned from the tanks twice daily until little more is produced, indicating that all material has been voided from the digestive tracts. In larger animals, it is preferrable to excise the digestive tracts soon after collection rather than allowing the animals to excrete their contents. Removal of ingested materials from the animals eliminates material which may contain inert constituents and contaminants in forms which do not become biologically available during passage through the gut. The shells of molluscs and exoskeletons of crustaceans are also excluded from analysis since these structures generally contribute considerable weight but low levels of contaminants.

The chemical constituents to be assessed for bioaccumulation are those deemed critical after considering known inputs from the sediment to be dredged. These constituents include organohalogens, mercury, cadmium, petroleum hydrocarbons and "known or suspected carcinogens, mutagens and teratogens".

If tissue concentrations in any of the dredged material samples are higher than in controls, analysis of variance and multiple range tests are used to identify significant differences. Peddicord (1977) recommended that any significant differences between tissue levels in test and control animals warrant potential cause for concern. Regulatory authorities must then objectively consider the magnitude of bioaccumulation shown, the toxicological significance of the materials accumulated, the proportion of the sediment sampling sites which produce uptake, the number of constituents bioaccumulated, the position of the species showing uptake in human and non-human food webs, the presence of motile species which might serve to transport bioaccumulated materials from the disposal area, and other factors relevant to the particular disposal operation in question.

Laboratory assessments of bioaccumulation potential should be conducted only in cases where true historical precedents for the proposed operation do not exist (Peddicord 1977). Animals surviving solid or suspended particulate phase bioassays may be used for laboratory assessments of bioaccumulation. Methods for conducting bioassays were outlined by Shuba and Tatem (1977) and Swartz et al. (1977) as summarized in Section 6. Survivors from the replicate controls are treated in a manner to correspond with reference samples from outside of the disposal site in the field assessment outlined earlier. Animals from the replicate sediment exposure aquaria correspond to samples from the disposal site in the field assessment outlined earlier. Survivors from the first replicate of all test medium concentrations in suspended particulate bioassays are pooled to make one sample corresponding to a disposal site sample; survivors from the second set of all test medium concentrations are pooled to make the second disposal site sample, etc. At the end of the bioassays, animals are placed in clean, sediment-free water to void the digestive tracts, as discussed earlier.

Tissue analyses and statistical tests are conducted as described for field assessment procedures.

RECOMMENDATIONS FOR THE DEVELOPMENT OF PROCEDURES FOR MONITORING BIOACCUMULATION OF CONTAMINANTS FROM GREAT LAKES SEDIMENTS

It is recommended that procedures followed in the United States for assessing the potential for bioaccumulation of contaminants from dredged materials disposed in ocean waters (U.S. Environmental Protection Agency/Corps of Engineers 1977) be examined for their adaptability for monitoring the bioaccumulation potential of contaminants in Great Lakes sediments.

Authorities with the U.S. Environmental Protection Agency and the Ocean Dumping Control Program of the Environmental Protection Service should be contacted to determine whether new techniques or refinements of existing techniques have been developed for investigating the bioaccumulative effects of sediments. The suitability of these techniques for biomonitoring of Great Lakes Sediments should then be assessed.

As detailed by Peddicord (1977), field assessments of biological uptake are preferable to laboratory evaluations. Uptake in the field should be monitored where true historical precedents for proposed dredging and disposal operations exist. Tissue levels of contaminants should be measured in animals collected from at least three stations within the disposal site boundaries, from three stations outside of the disposal area but in the path of sediment transport from the disposal site, and from three stations in a direction opposite that of net bottom transport. Caged animals may be required where a suitable species for monitoring uptake does not occur throughout the study area. Tissue levels in animals collected at dredging sites may be poor indicators of bioaccumulation potential of sediments since water contamination is also often severe in these areas and significant uptake from the water column should also be suspected.

Laboratory tests for determining the bioaccumulation potential of sediments are recommended where conditions required for field assessments are not met (Peddicord 1977). In this case, tissue contaminant levels should be compared in animals surviving solid phase bioassay tests (see Section 6).

Investigations should be carried out to provide a list of animals native to the Great Lakes that are suitable for monitoring the bioaccumulation potential of sediments. Species selected for analysis in field assessments should be relatively immobile so that contaminant body burdens reflect local environmental conditions. Some of the species proposed for use in sediment bioassays may also be suitable for bioaccumulation testing. The amphipod Pontoporeia affinis is ubiquitous in deeper areas of the Great Lakes (Cook and Johnson 1974) and may be a suitable species for use in field assessments. Unionid clams are useful organisms for monitoring bioaccumulation (Merlini et. al. 1978) and their potential for use in biomonitoring of sediment quality in the Great Lakes should be investigated further. Sphaeriidae and Chironomidae are also relatively abundant in the Great Lakes and may be useful biomonitoring organisms. Isopods are relatively tolerant of sediment contamination (Prater and Anderson 1977; Shuba et al. 1978) and may, therefore, be suitable organisms for determining bioaccumulation potential in field or in laboratory assessments of Great Lakes sediments. The fathead minnow, Pimephales promelas, is also relatively insensitive to sediment contamination (Prater and Anderson 1977) and readily accumulates contaminants from sediments (Halter and Johnson 1977). It is recommended that the use of fathead minnows in laboratory studies of the bioaccumulation potential of Great Lakes sediments be examined further.

While oligochaetes are ubiquitous in the Great Lakes, their small size may preclude them for use in bioaccumulation monitoring.

SEDIMENT BIOASSAY TECHNIQUES

Bioassay is defined as "any test in which organisms are used to detect or measure the presence or effect of one or more substances or conditions" (U.S. Environmental Protection Agency 1973). A bioassay is comprised of three components: 1) a stimulus such as a drug, pesticide or industrial waste; 2) a subject, which may be a cell, a tissue, or an organism: and 3) the subject's response (Alderdice 1967).

Relatively few bioassays have been conducted to monitor the effects of contaminated sediment. Two general types of sediment bioassays may be carried out. One type is concerned with water column effects and the other type with the effects of the solid phase. Water column toxicity has been monitored using both sediment elutriates and sediment suspensions. Engler (1979) reviewed field methods, laboratory tests, and toxicity data relating to toxicant bioaccumulation from contaminated sediments. Recently sensitive and rapid bioassay techniques have been developed at the Canada Centre for Inland Waters using different size fractions of phytoplankton as test organisms and sediment elutriate (Munawar _et_ _al_. 1983a).

WATER COLUMN TOXICITY

The U.S. Environmental Protection Agency/Corps of Engineers (1977) stated that the liquid phase (elutriate) of dredged materials may be evaluated for potential environmental impact either by chemical analysis and comparison with water quality criteria or by bioassays. The suspended particulate phase of dredged materials may only be evaluated by bioassays. Bioassays of each phase must test at least three species consisting of one phytoplankton or zooplankton species, one crustacean or mollusc, and one fish.

Phytoplankton, Zooplankton, Bacteria and Protozoa

Recommended U.S. Environmental Protection Agency Procedures

U.S. Environmental Protection Agency guidelines for conducting liquid phase phytoplankton bioassays to assess the toxicity of contaminated sediments proposed for ocean disposal on natural phytoplankton communities were outlined by Shuba et al. (1977a). In the author's opinion, the effects on phytoplankton at disposal sites are minimal due to the dynamic and variable nature of phytoplankton communities and because of rapid mixing and dilution occurring in the water column.

Phytoplankton bioassays with suspended particulates are not recommended due to interference and predation by indigenous protozoa and to interference by suspended particulates in the determination of response. Instead, Shuba et al. (1977a) suggested that zooplankton bioassays on suspended particulates be carried out.

A series of treatments and controls are set up using the sediment liquid phase and filtered disposal site water. Axenic cultures of phytoplankton from the disposal site are established or cultures of recommended species are acquired from suppliers. Procedures for algal assays for marine disposal sites are similar to those outlined by the U.S. Environmental Protection Agency (1974). Test organisms are inoculated into a series of concentrations of liquid phase (up to 100 percent). Nutrient additions may be required to distinguish growth inhibition due to toxicity and due to a lack of required nutrients. Observations are made after 4, 24, and at 24 hour intervals thereafter. Assays are continued until maximum cell numbers occur in each treatment. Cell density, cell volume, C^{14} uptake or chlorophyll concentrations may be used as response criteria.

Since phytoplankton data are estimates of population response rather than discrete counts of individual responses, as in animal bioassays, the statistical analyses described for animal bioassays are not applicable. The effective concentrations causing 50 percent inhibition (EC50) of the sample relative to the controls are calculated as outlined by Shuba et al. (1977a).

The evaluation of sediment quality must consider the amount of on-site mixing and dilution with the water column at the disposal site. Such an analysis is necessary to determine limiting permissible concentration of contaminants by means of phytoplankton bioassays.

Plankton/elutriate bioassays procedures followed at the Canada Centre for Inland Waters.

The experimental design for phytoplankton and zooplankton adopted at the Canada Centre for Inland Waters, Burlington, Ontario, Canada is shown in Figures 2 and 3 and is self explanatory.

Algal Fractionation Bioassays (AFB) and species composition

The experimental protocol for the algal fractionation bioassays to assess the impact of sediment-associated contaminants on different size fractions of phytoplankton is shown in Figure 3. In these experiments indigenous natural phytoplankton populations or communities collected from oligotrophic/eutrophic environments are exposed to increasing dosages of sediment-elutriates and their photosynthetic uptake is measured by means of Carbon-14 uptake technique (Vollenweider, 1969; Munawar et al., 1978). A portion of the well-mixed phytoplankton sample which is used as a test assemblage is preserved in Lugol's solution for taxonomic identification/enumeration and size analyses (Munawar et al., 1974; Munawar & Munawar, 1978). The remaining water sample is used for the Carbon-14 bioassays with different doses of the elutriate. The following experimental design is adopted from Munawar et al. 1983.

(a) dilution series consisting of four elutriate additions were run on each sediment with freshly prepared elutriates.

(b) two types of elutriate were used at each of the above concentrations: one which was passed through Chelex-100 resin (to remove dissolved metal ions) and the other the standard elutriate which contained nutrients as well as dissolved metals.

The C^{14} algal fractionation bioassays (AFB) were conducted using natural phytoplankton as a test population. The procedure is given below:

1. Phytoplankton test population is transferred to five 300 ml precleaned polycarbonite bottles fitted with lids.

FIGURE 2: ELUTRIATE/BIOASSESSMENT DESIGN

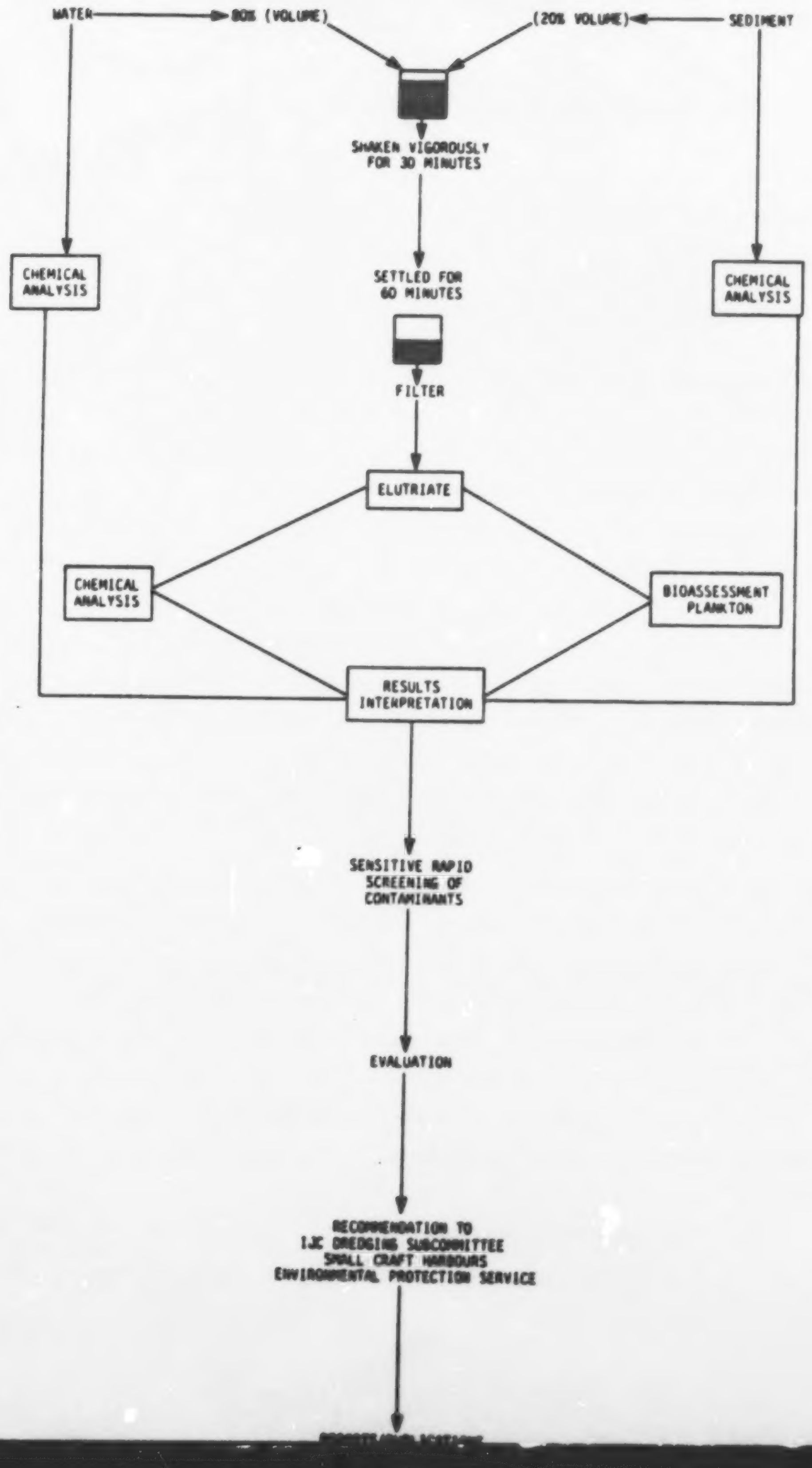
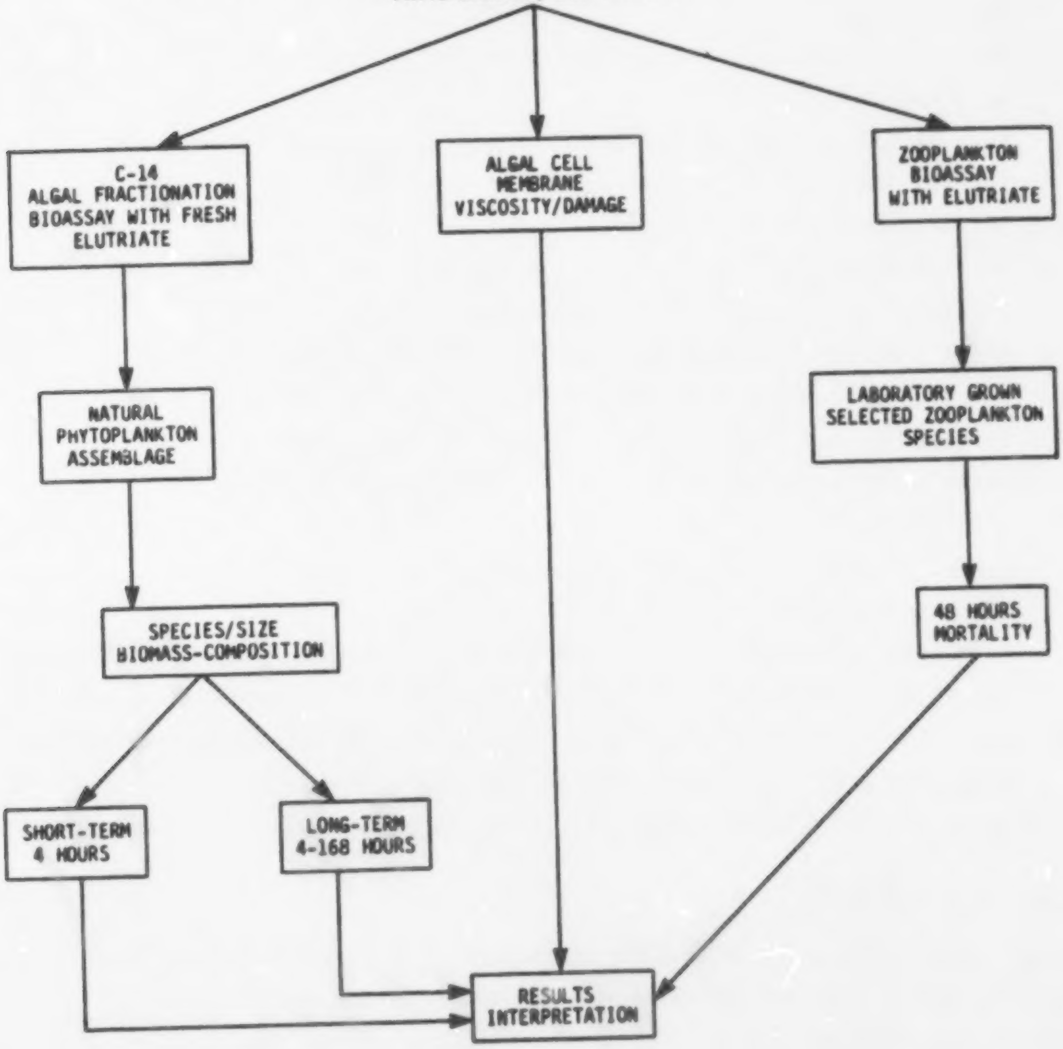


FIGURE 3: PLANKTON BIOASSESSMENT WITH ELUTRIATE PHYTOPLANKTON, ZOOPLANKTON


C-14 ALGAL FRACTIONATION BIOASSAY WITH FRESH ELUTRIATE
ALGAL CELL MEMBRANE VISCOSITY/DAMAGE
ZOOPLANKTON BIOASSAY WITH ELUTRIATE
NATURAL PHYTOPLANKTON ASSEMBLAGE
LABORATORY GROWN SELECTED ZOOPLANKTON SPECIES
SPECIES/SIZE BIOMASS-COMPOSITION
48 HOURS MORTALITY
SHORT-TERM 4 HOURS
LONG-TERM 4-168 HOURS
RESULTS INTERPRETATION

2. To these bottles various doses of elutriate (5, 10, 20 and 40 percent) were added as a source of sediment-associated contaminants.
3. One of the bottles to which elutriate was not added served as a control.
4. Four replicates of control and each elutriate concentration (Chelex 100 treated and untreated) were prepared.
5. The test bottles were spiked with 600 µl of $NaH^{14}CO_3$ and incubated for four hours at constant light in a laboratory incubator maintained at lake temperature.
6. After the four-hour incubation, 50 ml of the sample in triplicate from each bottle (treated and untreated) were filtered directly through a 0.45 µm Millipore membrane to estimate total primary production. Aliquots of 50 ml in triplicate from the control and elutriate-treated bottles were passed through a 64 µm, 20 µm and 5 µm Nitex screens and subsequently through a 0.45 µm Millipore membrane filter. The filters were then acidified with 10 ml of 0.1N HCl and kept in a Phase Combining System (PCS) for liquid scintillation counting (Lind and Campbell, 1969). Details of this algal fractionation bioassay (AFB) procedure are given in Munawar and Munawar (1982), which is a new technique used to assess the differential response of algal size assemblages to contaminants stress.

Zooplankton bioassays

Selected species of zooplankton either indigenous or collected from oligotrophic/eutrophic environments and grown in laboratory conditions are exposed to increasing dosages of elutriates (Munawar, unpublished report). All test organisms are kept in aquarium cultures in an incubator at constant conditions of light and temperature. At least fifty organisms were tested in each of the doses. For each concentration tested, the animals are divided into five replicates of ten individuals and placed in 250 ml polymethylpentene beakers containing 200 ml of test solution (filtered lake water plus elutriate). The experimental containers are not manipulated and the animals are not fed during the tests. Each test is conducted for a period of 48 to 96 hours, with all replicates at each concentration run simultaneously with one control. Observations of mortality are made within a few minutes under a stereomicroscope. The criteron for death is the failure to move when gently prodded by the tip of the micropipette.

The tests are performed under constant conditions of temperature (18 ± 1°C), lighting (white fluorescent tubes) and photoperiod (16 hrs. light/ 8hrs. dark). These conditions are similar to those recommended by the United States Environmental Protection Agency (U.S. EPA, 1978). A computerized probit analysis is carried out after (Finney, 1971). Mortality curves are drawn and where possible LC-50 values are calculated.

Other Procedures

A study-by-study account of published toxicity studies using micro-organisms follows.

Hendricks (1971) used a 0.3 M phosphate buffer at pH 7.0 to elute loosely bound chemical constituents from river sediment. The buffer elutriates contained protein, ammonia, and hexose sugars in concentrations of four to six times greater than found in river water or in equivalent elutriates using river water instead of buffer. He then measured the respiration rate of pathogenic and non-pathogenic enteric bacteria in these elutriates and demonstrated that the phosphate elutriate stimulated respiration rates in comparison with respiration measured using river water.

Shuba et al. (1977b) used freshwater and marine algal bacteria and protozoans to assess biological responses to elutriates of various harbours including Ashtabula Harbor, Lake Erie. Results of algal assays indicated both stimulatory and inhibitory effects of various sediment elutriates when growth in the elutriate was compared with growth in filtered disposal site water over periods of 8 to 18 days. Growth of bacteria and growth and respiration of protozoans in elutriates were also monitored but results were difficult to interpret because nutrient growth media usually had to be added to elicit a measurable response. The authors encountered difficulties in measuring the effects of potential contaminants, apparently due to the stimulatory effects of the nutrient media.

Hannan et al. (1976) elutriated coastal sediments from Washington State and California with artificial seawater and conducted bioassays on the elutriates with the marine algal, Phaeodactylum tricornutum. Two concentrations of elutriates were used for each sediment and each test and control solution received an equal inoculum of cells and the same concentration of added nutrients. Uptake of CO_2, cell densities, and biomass were determined after 48 hours. The authors felt that their technique provided a useful tool in classifying sediments according to toxicity and suggested that algal toxicity tests be carried out in combination with other bioassays for the evaluation of sediment quality.

In a more recent study, Hannan and Patouillet (1979) monitored the growth rate of Phaeodactylum tricornutum in elutriates of sediments collected from a diversity of marine and estuarine sites. Cultures were maintained in combinations of growth media and elutriates and growth rate was monitored over 20 to 44 hours by fluorescence. Although few cases of acute toxicity were found, elutriates often induced a prolongation of the lag phase of growth. Results showed good reproducibility.

Gannon and Beeton (1969, 1971) conducted algal assays on selected dredged sediments from Great Lakes harbours. The authors measured uptake of C^{14} and also attempted direct cell counts and light transmittance measurements to determine cell densities in cultures maintained in unfiltered sediment extracts. Direct cell counts and light scattering methods were unsuccessful due to the aggregation of algal cells with sediment particles. Assays with C^{14} indicated that C^{14} uptake and percent sediment extract were directly related over a four-day experimental period.

Flint and Lorefice (1978) performed in situ primary productivity bioassays with elutriates of Ashtabula Harbor (Lake Erie) sediments. The dilution water used was filtered lake water. Bioassays were conducted using a series of elutriate concentrations with phytoplankton samples collected from the lake. Light and dark bottles were then suspended in the lake at a depth of 1 m and C^{14} uptake was monitored over four hours. The experiment was repeated four times from May to September using samples of the natural phytoplankton community found in the lake on each occasion. Analysis of variance was used to show that photosynthesis is significantly inhibited at higher elutriate concentrations. The authors pointed out that, unlike some algal assay techniques which require the monitoring of population changes over days or weeks, their procedure is relatively simple and provides informative results in a short period of time. Furthermore, the short duration of this test was considered representative of the immediate impacts typical of dredging operations.

Macroinvertebrates and Fish

Recommended U.S. Environmental Protection Agency Procedures

U.S. Environmental Protection Agency guidelines for performing liquid phase and suspended particulate phase animal bioassays were outlined by Shuba and Tatem (1977). These bioassays were intended for use in the evaluation of the impact of suspended particulates and dissolved chemicals released from sediment during ocean disposal operations in U.S. waters.

Appropriate sensitive test species are selected as described in Appendix A of U.S. Environmental Protection Agency/Corps of Engineers (1977) guidelines for the ecological evaluation of proposed discharges of dredged materials into ocean waters. When possible, test species should be collected from an area near the disposal site with similar water and sediment quality. If this is not possible, species phylogenetically or ecologically similar to the dominant sensitive species expected at the disposal site are selected. All liquid and suspended solid phase bioassays should include a mysid shrimp of the genus Mysidopsis or Neomysis to provide an internal standard and form a basis for quality assurance in the regulatory program. Juvenile fish are generally more sensitive and are preferred over adults. Wet weights of individual organisms should not exceed 3 grams. Molluscs are generally resistant and therefore, may not be suitable for acute toxicity work. When used, molluscs should be less than 2 cm long.

Procedures for the collection and preparation of disposal site water and dredged material samples were described in Appendix B of the U.S. Environmental Protection Agency/Corps of Engineers (1977) guidelines. During bioassays, temperature, salinity, dissolved oxygen, and pH should be monitored and reported daily. Temperatures should be maintained within 2°C of the temperature expected at the disposal site. Dissolved oxygen should not be permitted to fall below 4 mg/l unless lower values are expected to occur for a substantial time during disposal or if lower values occur naturally at the site.

Tests are carried out using 200 ml of liquid in crystallization dishes (for zooplankton or larval fish) or 30 L of liquid in glass aquaria (larger animals).

A minimum of three replicates of each experimental and control condition is recommended. Ratios of at least three concentrations of test liquid (elutriate or suspended solids phase) - 10, 50 and 100 percent - in disposal water are recommended. Ten organisms are exposed in each replicate for 96 hours. The test media are not replaced and aeration is not supplied unless deemed necessary due to low oxygen conditions. Observations should be made at intervals of 0, 4, 8, 24, 48, 72 and 96 hours. If more than ten percent mortality occurs in the controls, the data are discarded and the experiment must be repeated. Control mortalities of 20 percent may be acceptable in zooplankton and larval bioassays. Liquid and suspended particulate phase bioassay results are interpreted in context with the mixing and dilution expected at the disposal site. Dredge disposal is only environmentally acceptable if bioassay results and initial mixing estimates indicate that the limiting permissible concentration will not be exceeded. Detailed information on statistical analysis of results and data interpretation is also provided.

Shuba et al. (1978) conducted numerous bioassays in which freshwater and marine invertebrates were exposed to heavily contaminated sediments, sediment elutriates, and suspended sediments to further refine biological methods for assessing the effects of open-water disposal of dredged materials on biota. Freshwater and marine sediments representing a wide variety of sediment-contaminant combinations were collected from various sources in the United States. Marine test animals included: *Acartia*, *Mysidopsis*, *Palaemonetes* (*P. pugio*), *Neanthes*, *Rangia*, *Mercenaria*, and benthic amphipods and isopods. Freshwater species were represented by : *Palaemonetes* (*P. kadiakensis*), *Daphnia*, *Corbicula*, *Musculium* and *Lirceus*. Most tests were short-term acute bioassays. Only one sediment was shown to be clearly toxic to a majority of the animals tested. Many sediments proved to be non-toxic, despite high contaminant concentrations. One long-term bioassay with grass shrimp larvae illustrated the potential of contaminated sediments for causing chronic effects on growth and reproductive capacity.

From these studies, Shuba et al. (1978) recommended that physicochemical conditions (dissolved oxygen, pH, salinity, temperature) should be closely monitored during toxicity testing. Sediments should be stored for only short periods and should be refrigerated before testing due to loss of toxicity during storage. Of the animals tested, the following marine organisms were recommended for further bioassay work: *Acartia tonsa*, *Sphaeroma quadridentatum*, *Mysidopsis* sp., *Parahaustorius* sp., and larval *Palaemonetes pugio*. *Trigriopus californicus*, *Rangia cuneata* and *Mercenaria mercenaria* were pollution-tolerant and not recommended for acute toxicity work. The freshwater species *Palaemonetes kadiakensis*, *Daphnia pulex* and *Musculium* sp. were recommended as test species, while *Lircus* and *Corbicula* were highly tolerant and not recommended for acute toxicity studies. Other recommended procedures included the preference for flow-through rather than static systems and the determination of reproducibility by performing replicate experiments.

Other Procedures

A study-by-study account of liquid and suspended particulate phase bioassays using macroinvertebrates and fish follows.

DeCoursey and Vernberg (1975) collected water samples from upland disposal area runoff, from 200 yards downstream of a dredge site during dredging and disposal operations in Charleston Harbor, South Carolina. Bioassays were conducted with the collected water on juvenile water fleas (*Daphnia pulex*), larval grass shrimp (*Palaemonetes pugio*) and larval polychaetes (*Polydora*).

Survival, respiration rate and swimming rate were measured to assess acute and chronic toxicity. Disposal site runoff water was most toxic by all response criteria.

Lee et al. (1975) used the freshwater cladoceran, *Daphnia magna* and the marine shrimp, *Palaemonetes pugio* in static bioassays of many sediments from the United States, including one from Ashtabula Harbor, Lake Erie. Elutriates were prepared by mixing various percentages of sediment (5, 10, 15 and 20 percent) with water using compressed air. In some cases, growth media were used rather than disposal site water to prepare the elutriates. Some elutriates were acutely toxic in 96-hour tests. The authors recommended the use of benthic organism bioassays for the assessment of the effects of dredged sediment disposal.

A series of sediment bioassays were carried out on Los Angeles Harbor sediments through the University of Southern California's Institute of Marine and Coastal Studies (Soule and Oguri, 1976).

Emerson (1976) used two benthic polychaetes, *Ophryotrocha* sp. and *Capitella capitata* to assess the toxicity of elutriates of Los Angeles Harbor sediments. Elutriates were filtered to remove all suspended particulates. Survival in a static bioassay system was then monitored. Mortality of *C. capitata* was less than 50 percent while there were no mortalities of *Ophryotrocha* sp. in 96-hour tests. Long-term (28-day) tests were also conducted to determine the effects of sediment elutriates on reproduction and developmental success. *Ophryotrocha* sp. produced significantly fewer numbers of offspring in most elutriates. Survival of *C. capitata* larvae was reduced in elutriates of more grossly contaminated sediments, although higher growth rates were exhibited by survivors in these elutriates.

Brewer (1976) prepared elutriates of sediments from three areas of Los Angeles Harbor to assess their toxicity to juvenile and adult northern anchovy, *Engraulus mordax*. Elutriates were prepared from seawater-sediment ratios of 4:1 to 100:1. Solids were allowed to settle for 60 minutes but elutriates were not filtered or centrifuged. Fish were exposed to aerated elutriates in 96-hour tests and for longer periods of up to 14 days. Results were inconclusive, apparently due to hypoxic conditions in the elutriates obscuring any toxicity effects.

Chamberlain (1976) exposed the California killifish, *Fundulus parvipinnis* and white croaker, *Genyonemus lineatus* to centrifuged elutriates of Los Angeles Harbor sediments in 96-hour bioassays. No substantial mortality occurred in short-term tests. Long-term exposures (up to 28 days) were also carried out but the author felt that deaths of test animals reflected starvation rather than toxicity.

McConaugha (1976) subjected two species of copepods, *Acartia tonsa* and *Tisbe* sp. to filtered elutriates of sediments from 12 sites in Los Angeles Harbor. Toxicity tests were performed over 96-hours in one concentration (100 percent) of elutriates prepared using sediment-seawater ratios of 1:4. Increased mortalities in elutriates compared with controls occurred for certain sediments while in other elutriates, survival was significantly higher than in controls. Elutriates of sandy sediments with relatively high levels of heavy metals and chlorinated hydrocarbons were non-toxic while all sediments yielding toxic elutriates were relatively fine, suggesting that finer grain sizes may have allowed greater release of toxins.

Gannon and Beeton (1969, 1971) performed short-term, acute bioassays on benthic fauna (*Pontoporeia*, *Gammarus*, larval *Chironomus*), *Daphnia* and native zooplankton using suspensions of sediments from Great Lakes harbours. Data from these tests were inconclusive since dissolved oxygen measurements were not taken and the authors suspected that the high oxygen demand of some sediments rather than toxic sediment constituents may have contributed to mortality.

Peddicord and McFarland (1978) studied the toxicities of suspensions of relatively uncontaminated sediments and contaminated sediments from the Sacromento River-San Francisco Bay system. Bioassays were conducted over 21 days in suspended sediment concentrations of 2 to 20 g/l. Test organisms included several marine, estuarine, and freshwater species. The freshwater species used were a clam, *Corbicula fluminea*, an odonate, *Enallagma* sp., an amphibian, *Bufo boreas*, a cyprinid, *Notemigonus crysoleucas*, and a salmonid, *Salmo gairdneri*. Exposure to contaminated sediments decreased survival substantially over exposure to uncontaminated sediments.

Legore and Desvoigne (1973) carried out bioassays with suspensions of sediment from the Duwamish Waterway of Seattle, Washington. Doses of up to five percent by weight elicited no apparent response from threespine sticklebacks (*Gasterosteus aculeatus*) or coho salmon fry (*Oncorhynchus kisutch*) in static 96-hour tests.

Tsai et al. (1979) carried out a series of bioassays on contaminated Baltimore Harbor sediment using mummichogs (*Fundulus heteroclitus*), spot (*Leiostomus zanthurus*) and clams (*Mya arenaria*). Test animals were exposed in static systems to a series of suspended sediment concentrations, to Fuller's earth suspensions (controls) or to uncontaminated test seawater for 48 hours. Twenty-four and 48-hour TLM's (Tolerated Lethal Median) with sediment concentration. Mummichog 24 h TLm data from sediments collected at various harbour sites permitted the delineation of zones of toxicity in the harbour.

SOLID PHASE TOXICITY

Acute Toxicity

Recommended U.S. Environmental Protection Agency Procedures

The U.S. Environmental Protection Agency/Corps of Engineers (1977) requires that solid phase bioassays be carried out with appropriate sensitive benthic marine organisms in the technical evaluation of proposed ocean disposals of dredged materials. The solid phase is not mixed or dispersed as quickly as the liquid and suspended particulate phases and benthic organisms may be exposed for extended periods following disposal. Solid phase bioassays must use at least three species including a filter-feeder, a deposit-feeder and a burrowing species.

Results of experiments that contributed to the development of the U.S. Environmental Protection Agency's standard techniques for performing solid phase bioassays to assess the toxicity of sediment proposed for ocean disposal were described by Swartz et al. (1979). Toxicity tests on contaminated sediments from U.S. coastal areas were conducted using five macrobenthic invertebrates including the bivalves Protothaca staminea and Macoma inquinata, the polychaete Glycinde picta, the amphipod Paraphoxus epistomus and one of four cumacean species. The technique involved dumping a layer of test sediments onto benthic organisms established in a layer of disposal site or reference sediments in aquaria. Mortalities in various depths of test sediment were monitored to determine an optimum test sediment depth of 15 cm. Survival in test sediments relative to controls after ten days of exposure was the response criterion. Crustaceans were consistently more sensitive to contamination than either polychaetes or molluscs.

The U.S. Environmental Protection Agency guidelines for conducting solid phase bioassays to assess the toxicity of contaminated sediment to benthic organisms were described by Swartz et al. (1977) as outlined in the following paragraphs.

Based on the premise that conditions within the disposal site are expected to be adversely affected by disposal while conditions beyond the disposal site should not be affected, the objective of solid phase bioassays is to determine the potential impact of the solid phase on benthos at and beyond disposal site boundaries.

Appropriate test species selected from Appendix A of U.S. Environmental Protection Agency/Corps of Engineers (1977) are collected from the disposal site area. As previously mentioned, these include a filter-feeder, a deposit-feeder and a burrowing species. Swartz et al. (1977) further recommended that these include a crustacean, an infaunal bivalve and an infaunal polychaete. Infaunal amphipods are among the most sensitive crustacea and are preferred for solid phase bioassays. Also, bioassays should include a mysid shrimp of the genus Mysidopsis or Neomysis to provide an internal standard and form a basis for quality assurance in the regulatory program. If the dominant species near the disposal site cannot be used, guidance is given for selecting phylogenetically or ecologically similar species for toxicity testing. Juvenile forms, particularly of molluscs and large crustaceans, are generally more sensitive than adults and are therefore preferred.

Reference sediment is collected from the vicinity of the proposed disposal site and test sediment is collected from the proposed dredge site. Bioassays are carried out on sediments from at least two areas of the proposed dredging site if heterogeneity of sediment quality is expected. Sediments are wet-sieved to remove organisms and are stored at 2 to 4°C for no longer than two weeks before use.

Five replicate aquaria are established for each test sediment and for controls. More replicate aquaria may be required if predator-prey relationships occur among the test animals. Seawater of similar physicochemistry to disposal site water is used in the test. A flow-through system that initially passes water through a 20 um pore size filter and replaces the aquarium volume at least once every 4 hours is preferred, although static water systems may be used as long as 75 percent of the water volume is replaced every 48 hours.

A 30 cm layer of reference sediment is placed on the bottom of each aquarium and allowed to settle for 1 hour. Twenty individuals of each test species are placed in each aquarium and are allowed to acclimate for 48 hours. Dead animals are removed and replaced after this period. Test sediment is then added to the test aquaria to establish a 15 cm layer above the reference sediment. A 15 cm layer of reference sediment is added to controls. Some crustaceans such as mysid shrimp may not survive sediment addition and must be introduced immediately after sediment is added.

The bioassay is continued for ten days and daily observations are made of obvious mortalities and unusual occurrences. Salinity, dissolved oxygen, and temperature are also recorded daily. Gentle aeration may be used to maintain oxygen levels above 4 mg/l, unless lower concentrations are expected for a substantial time at the disposal site. After ten days, the sediments are sieved through a 0.5 mm screen and mortality is recorded. An average control mortality of greater than ten percent is unacceptable and necessitates the repetition of the bioassay.

Analysis of variance and multiple range tests are then carried out on the data to detect significant differences among mortalities in the various test and control sediments. If the difference between mean survival in test and control animals is significant and exceeds ten percent, the limiting permissible concentration is considered to be exceeded and the proposed disposal operation is deemed environmentally unacceptable.

Other Procedures

Other published accounts of acute solid phase bioassays are summarized in the following paragraphs.

Prater and Anderson (1977) conducted 96-hour sediment bioassays on eight sediments collected from Duluth and Superior Harbors, Lake Superior using the burrowing mayfly, Hexagenia limbata, the isopod, Asellus communis, the water flea, Daphnia magna and the fathead minnow, Pimephales promelas. Bioassays were conducted in aquaria with continually recirculating water over about 5 cm of test sediment. This circulation maintained an adequate oxygen supply in aquaria. Oxygen levels were determined daily. Mortality was determined after the 96-hour exposure period. Mortalities of <10 percent, 10 to 50 percent, and >50 percent were considered indicative of unpolluted, moderately polluted and heavily polluted conditions respectively. Daphnia magna and H. limbata were the most sensitive species to sediment contamination, while P. promelas was relatively insensitive.

Birge et al. (1977) conducted bioassays on control and metal-contaminated sediments from Kentucky streams using fertilized eggs of the narrow-mouthed toad, Gastrophryne carolinensis, goldfish, Carassius auratus and rainbow trout, Salmo gairdneri. Experimental vessels were 500 ml chambers containing two cm of test sediment and synthetic culture water. Gentle, continuous aeration was used to maintain oxygen levels near saturation. Using a sample size of 100, treatment was initiated at fertilization for the narrow-mouthed toad, two to four-hour post-fertilization for the goldfish and on the 15th day of development for trout embryos. Temperatures were maintained at appropriate temperatures for each species. Exposure was maintained through four to ten days post-hatching. Sediments were substantially more toxic to eggs and embryos than to free-swimming larvae or fry. Embryonic mortality was always higher in test sediments than in controls. Also, the frequency of teratogenesis in surviving populations was appreciably higher for test sediments than for controls. In trout, frequencies of mortality and teratogenesis paralleled sediment heavy metal concentrations. This correlation was not evident for toads and goldfish which have shorter sensitive embryonic periods. The authors concluded that such short-term hatchability tests were useful for the prediction of long-term ecological effects of aquatic and sediment-accumulated pollutants.

Chronic Toxicity

Accounts of solid phase chronic toxicity have also been published, as summarized in the following paragraphs.

Gannon and Beeton (1969, 1971) studied the selection of contaminated Great Lakes sediments by benthic organisms. The experimental animals used were the amphipods Pontoporeia and Gammarus and larvae of the midge Chironomus. Animals were placed in test chambers containing contaminated and control sediments and sediment preferences were monitored. These experiments showed that test organisms preferred some sediments over others. However, there were no clear relationships between sediment contamination and selection by the animals.

Wentsel et al. (1977) performed chronic toxicity tests to determine the effects of heavy metal contamination in sediment on growth and survival of the freshwater chironomid, Chironomus tentans. Early instar larvae were maintained for 17 days in sediments having various levels of heavy metal contamination. Growth was inhibited by heavy metal contamination and a linear relationship was observed between the square root of larval length and metal levels in the sediment.

Wentsel et al. (1978) also assessed the effect of metal-contaminated sediment on the emergence of *C. tentans*. Mature larvae were added to beakers containing control sediment or heavy-metal-contaminated sediment from an Indiana lake. Emerging adults were counted daily for 14 days after the first emergence. Approximately three times as many adults emerged from control sediments than from test sediments. Emergence was also delayed by two days in the contaminated sediment.

In a study using a marine species, McGreer (1979) monitored the chronic toxicity of contaminated sediment from the Fraser River estuary (British Columbia). The burrowing and avoidance responses of the bivalve, *Macoma balthica* were observed in sediments containing various levels of heavy metals. Each of five containers were filled with a different sediment. Ten clams were placed on each sediment and the number of animals which had burrowed were recorded at intervals of 15, 30, 45, 60, 90, 120 minutes and 6, 12, 24 and 48 hours. Burrowing behaviour was inhibited in all contaminated sediments relative to controls, with the unit of response being the time required for 50 percent of the population to burrow (ET50). Burrowing time was significantly related to sediment Hg and Cd levels. Clams also showed a significant avoidance of the most contaminated sediment when presented with a choice between a control and a polluted substrate. McGreer felt that these techniques provide a sensitive, simple and rapid biomonitoring tool.

Table 1: Species known from the Great Lakes showing promise for use in acute bioassays with sediment

Organism	Source
Crustacea	
Palaemonetes kadiakensis	Shuba et al. (1978)
Daphnia pulex	Shuba et al. (1978); DeCoursey and Vernberg (1975)
Insecta	
Hexagenia limbata	Prater and Anderson (1977)
Mollusca	
Musculium sp.	Shuba et al. (1978)
Pisces	
Salmo gairdneri embryos	Birge et al. (1977)

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